Particle Selection Using CsI(Tl) Crystals and Associated Experiments on the Luminescence of Alkali Halide Crystals.

by

J.C. Robertson

Department of Natural Philosophy,

University of Glasgow.

Presented as a Thesis for the degree of Ph.D. in the University of Glasgow, April 1961.

Preface.

The work described in this thesis was carried out during the period May, 1958 - September, 1960 in the Department of Natural Philosophy, The University of Glasgow.

The thesis is presented in two parts.

In Part I - "Particle selection using CsI(Tl) crystals", an introductory chapter reviews the experimental difficulties encountered in experiments with fast neutrons (14 MeV) and the techniques, with their limitations, previously used to surmount these difficulties. In Chapter II a description is given of a novel form of scintillation counting in which different types of particle are distinguished by the decay time of the luminescence they produce in a CsI(Tl) crystal. The use of this technique in reducing the unwanted, but unavoidable, backgrounds produced in CsI(Tl), when it is used as the detector in neutron experiments, is illustrated in Chapter III. Also discussed in this chapter are the advantages this new technique has over previous methods for studying neutron induced reactions. Chapter III concludes with a brief discussion of typical experiments which are now made possible by its use; in particular n-α and n-np reactions at 14 MeV. It is stressed that the technique is perfectly general in application and could be used equally well to study other types of reaction such as γ-p, γ-α reactions.

The author was solely responsible for the development of this technique but wishes to acknowledge the supervision of Dr. A. Ward during the early part of it.

In Part II - "Associated experiments on the luminescence of alkali halide crystals", Chapter I reviews the interpretation of the scintillation process in alkali halide crystals, used as the phosphor in scintillation counters. Chapter II describes experiments on the decay of the luminescence in various alkali halide crystals for particles of different ionization density while Chapter III discusses experiments on the luminescence of CsI(Tl) and CsBr(Tl) as a function of temperature, and the luminescence of CsI(Tl) as a function of thallium concentration. Chapter IV gives an interpretation of these experiments; and the importance of the work in relation to Part I of the thesis is stressed.

The author is solely responsible for the interpretation of the experiments given in Chapter IV. Much of this work was performed with the assistance of Mr J.G. Lynch. The experiment on the luminescence of CsI(Tl) and CsBr(Tl) as a function of temperature was performed in collaboration with Mr J.G. Lynch and Dr. W. Jack.

I should like to thank Professor P.I. Dee for his interest and encouragement and for many helpful discussions.

April 1961. J.C. Robertson.

FOOTNOTE: Two papers on the work described in this thesis have been published. These are: (a) 'Particle Selection in Crystals of CsI(Tl)', by J.C. ROBERTSON and A. WARD, Proc. Phys. Soc., 1959, 73, 523, and (b) 'The Luminescent Decay of Various Crystals for Particles of Different Ionization Density', by J.C. ROBERTSON and J.G. LYNCH, Proc. Phys. Soc. 1961, 77, 751. A third paper entitled 'The Luminescence of CsI(Tl) and CsBr(Tl) as a Function of Temperature', by J.C. ROBERTSON, J.G. LYNCH and W. JACK has been submitted for publication in this journal.

Particle Selection using CsI(Tl) Crystals and Associated Experiments on the Luminescence of Alkali Halide Crystals.

Contents.

Part I.

Particle Selection Using CsI(Tl) Crystals.

Part I.

Introduction.

Chapter I.

I.1. The Scintillation Counter.

The scintillation counter is a familiar instrument for the detection of nuclear particles in all fields of nuclear research. Its essential features are a luminescent solid, liquid or gas which emits scintillations when bombarded by the particles to be detected, and a photo-electric multiplier tube which responds to the scintillations and produces electrical pulses at its output. The number of pulses produced determines the number of incident particles, while the magnitude of the pulse is proportional to, or has a simple relation to, the energy of the incident particle. The advantages that this method of detection has over other methods, such as Geiger-Muller gas counters, are the very rapid response time and the high energy resolution which can be obtained, which are very advantageous for coincidence measurements.

The first attempts to use a phosphor with a photo-electric multiplier seem to have been made by Blau and Dreyfuss (1945), while the first attempts to detect individual particles using a scintillation counter seem to have been by Curran and Baker (1944).

The three types of luminescent materials used in scintillation counters, apart from gases, are as follows:- (a) inorganic crystals (b) organic crystals (c) organic liquids and plastics. Organic crystals, liquids and plastics are used predominately in high energy nuclear physics since the decay time of the luminescence is very much faster than the decay time of the luminescence in inorganic crystals, and they are therefore better suited to the very fast coincidence techniques employed. In low energy nuclear physics, on the other hand, inorganic crystals are usually used since they have a more proportional energy response than the organic materials and in general better energy resolution can be obtained with them. These inorganic crystals are usually activated by an impurity to increase the efficiency for conversion of particle energy into luminescence. The most commonly used inorganic crystals are the thallium activated alkali iodides which were developed by Hofstadter (1948).

In Part I of this thesis, a description is given of a novel form of scintillation counting. In order to bring out the importance of this work it is necessary to consider how the scintillation counter can be used to distinguish one type of particle from another and also to review the

other methods of particle detection commonly used. We limit ourselves to a consideration of the techniques used in low energy nuclear physics. In particular, to a consideration of the techniques used in experiments with fast neutrons (14 MeV) since the technique described in this thesis was developed primarily to permit the measurement of the energy spectra of particles emitted in n-α and n-p nuclear reactions at 14 MeV. Before discussing these methods of detection, however, it is convenient first to review the experimental difficulties encountered in making such measurements.

I.2. General Considerations of the difficulties encountered in experiments with fast neutrons.

The measuremont of the energy spectra of particles emitted in nuclear reactions induced by fast neutrons from the reaction $^{3}He(d,n)^{4}He$ (Q = 17.577 MeV) is rendered difficult because of (a) the low flux of neutrons available and (b) the backgrounds produced in the detector from nuclear reactions induced by the neutrons in the detector and its immediate surroundings.

In the experiments, in which we are interested, the source of neutrons is usually a tritium-titanium target provided by Harwell, in which 1/3 c.c. of tritium gas is absorbed into a layer of titanium ($\approx$ 200μ gms/cm^2 thick)

evaporated on to a layer of copper. To a beam of deuterons of energy 300 KV. this target is 70 KV. thick. An extremely mono-energetic source of neutrons is produced, approximately isotropic in distribution. A flux of 5×10^9 neutrons into 4π can be obtained from such a target for approximately ten hours, with an average beam current of approximately 20 μ.amp. When this is compared with the directed beam of 6×10^{12} charged particles/sec. obtainable in accelerators, with a beam current of 1 μ.amp, it is obvious that the chief problem in neutron experiments will be the detection efficiency. This flux of 5×10^9 neutrons into 4π is the maximum obtainable from such tritium-titanium targets. The flux cannot be increased by increasing the deuteron beam current since the heat dissipated then tends to 'burn up' the tritium-titanium target.

This problem of counting efficiency is also related to the low cross sections for the neutron induced reactions and the thin foils of target materials which must be used. The medium weight elements have n-p cross section ranging from 100-500 mb in value (Paul & Clarke, 1953), and are the most convenient to study. In the heavier elements, the Coulomb barrier has a dominating influence, and the n-p cross sections are in the range 1-10 mb. (Coleman et al., 1959). Due to

the greater Coulomb barrier, the values for the n-α cross sections would be expected to be correspondingly smaller than the values for the n-p cross sections. Paul and Clarke (loc. cit.) find values in the range 20-50 mb. for n-α cross sections in medium weight elements and values in the range 50-200 mb. for light weight elements. Coleman et al. (loc. cit.) find n-α cross sections in heavy elements to have values in the range 0.3-3 mb. It can be seen from these values of the n-p and n-α cross sections that the yield from such neutron induced reactions would accordingly be small. It is not possible to increase the yield by increasing the thickness of the target foils since then the energy resolution of the emitted particles would be impaired by the varying energy loss of the particles leaving the target foil.

The extreme penetrability of fast neutrons is illustrated by the fact that about 20 cms. of copper or steel are necessary to attenuate the flux by a factor of twenty. This means that the detector and its surroundings are always irradiated to some extent by fast neutrons which result in unavoidable backgrounds being produced. This is particularly serious in detecting particles of energy less than 4 MeV due to the very high flux of low energy β and

rays produced. It is necessary, of course, that the number of background events should be much less than the number of events in which we are interested. The signal to background ratio can be improved by arranging the geometry of the experiment so that the flux of neutrons at the target foil is greater than that at the detector by an appreciable factor. This method has been employed in both nuclear emulsion (Allan, 1957) and scintillation counter techniques (Colli et al., 1956). The background in the detector can be kept to a minimum by the use of very heavy elements such as Pb, Bi or Au in the detection arrangement. Carbon with an extremely negative n,p Q value (-12.59 MeV) is also a good background material with the additional advantage over the heavier elements that not so many β and γ rays are produced when it is irradiated with fast neutrons. However its mechanical properties, and the reaction $^{12}C(n,n')^{4}He$, Q = -7.28 MeV, limit its application. Although the background due to protons and alpha particles etc. may be reduced by the use of the materials mentioned, the undesirable background from β and γ rays remains. Finally we can screen the detector but this usually reduces the counting efficiency, and the effect of the flux of scattered neutrons from the room relative to the direct

flux from the tritium target must be considered.

In practice these methods prove fairly successful in obtaining a suitable signal to background ratio. In using scintillation counters, however, in which the minimum thickness of the crystal detector is determined by the maximum energy of the emitted particles, the low energy background from β and γ rays always remains. This low energy background begins at approximately 4 MeV and increases exponentially with decreasing energy.

I.3. The Experimental Techniques used in the study of the energy spectra of particles emitted in fast neutron reactions.

We now discuss the methods of detection employed in experiments with fast neutrons and how these methods are used to distinguish one type of particle from another.

I.3.a. The Nuclear Photographic Emulsion Technique.

In this technique the type of particle is determined by the grain density, measured as a function of the length of the track, produced by the particle in the emulsion. The energy of the particle is determined by the length of the track. The technique has been used successfully to measure n-p energy spectra (Brown et al., 1955; Allan, 1957/58/59; March & Morton, 1958a,b,c; Peck, 1957; Haling et al., 1957, Armstrong & Rosen, 1960; Ahn et al., 1960) and has much to

commend it. In particular the angular distribution of the reaction products can be obtained simultaneously with the energy spectra. It is also very easy to discriminate against the background from low energy β and γ rays with the result that the lower limit for detection is extended to 1 MeV compared with 4 MeV in counter experiments. Target foils in the form of separated isotopes can be used. To obtain a sufficient yield, however, it is necessary to use fairly thick targets with the result that at the lower proton energies, large corrections are necessary for the amount of energy lost by the protons in escaping from the target foil. In addition, the range of a low energy proton in the emulsion is very short, and considering the error in estimating the track length a greater percentage error will be present at the lower energies. Containing hydrogen, the photographic emulsion is at a disadvantage in fast neutron work due to the large number of recoil protons from the n,p scattering reaction in the emulsion. This difficulty can be overcome however by a suitable geometry or a shielding technique. The most serious criticism is the difficulty of obtaining good statistical errors due to the large time involved in scanning the plates. The best signal to background ratio obtained is about 3:1, and there is a fairly large statistical error in the spectra. For example, in a

typical experiment, such as that of March and Morton (1958) on Ni^{60}, 2498 tracks were examined of which 792 were due to background protons. It is the poor efficiency as well as the large scanning time which inhibits the application of this method to large numbers of elements. The method is also unsuitable for measurements on n-α spectra since the tracks produced by the alpha particles are exceedingly small and difficult, if not impossible, to distinguish from the tracks produced by low energy protons. Again there is the difficulty of obtaining good statistical errors. Kumabe et al. (1957) have in fact used this method to measure the n-α spectra from aluminium at 14 MeV.

I.3.b. Scintillation Counter Techniques.

Colli et al. (1956), and later Jack (1960), have used a scintillation counter telescope to measure n-p energy spectra at 14 MeV in medium weight elements. In this method of detection, the energy lost by a particle in a thin crystal, or proportional counter, is measured. This serves to identify the type of particle. The energy of the particle is then measured in a second crystal. A coincidence technique is then used. Only particles having a sufficient energy loss in the $\frac{dE}{dx}$ counter produce coincidences. In Colli's apparatus, proportional counters filled with (7 cms Hg) CO_2 were used to measure the energy loss while a thick CsI(Tl)

crystal was used to measure the energy. The counters were constructed from materials with a low n-p cross-section while the background produced in the CsI(Tl) crystal was kept to a minimum by arranging the geometry so that the neutron source was considerably nearer the target foil than the CsI(Tl) crystal. CsI(Tl) is a suitable choice as scintillator because of the low n-p cross sections of Cs^{133} and I^{124} (Coleman et al., 1959). With this method a notable improvement in the statistical error on n-p spectra are obtained. At low proton energies however, the background is determined by the random coincidence rate which due to the high flux of low energy β and γ rays is very large. This limits the lower energy limit to about 4 MeV and since a study of the results obtained using nuclear emulsions suggests, that in a typical n-p energy spectra, about 40% of the protons have energy between 2-4 MeV, this is quite a serious criticism. The angle of acceptance of the protons emitted from the target foil is approximately 20°-40° and the method is best suited for measuring the contribution to the proton energy spectra from direct interaction as opposed to compound nucleus effects. These would be expected to contribute mainly in the forward directions. Colli et al. were not able to use separated isotopes as targets due to the

rather large target areas required. This difficulty has been overcome by Jack (loc.cit.) who in addition obtained a smaller angle of acceptance and better energy resolution. Eubank et al. (1959a,b) have applied this technique to n-p measurements on heavier elements but the signal to background ratio was never greater than 1:2. The angular distribution of the protons is of course not obtained from a single run, separate runs being required at each angle.

Summarising, we can say that this method has proved very successful in measuring n-p energy spectra at 14 MeV. The method yields better statistics than the nuclear emulsions technique but has the disadvantage of being unsuitable for use in measuring the energy spectra below 4 MeV. It has not yet been applied to measuring n-α spectra at 14 MeV., although such an application might be possible.

Storey et al. (1960) have developed a technique for measuring the n-p energy spectra from medium weight elements, in which a single CsI(Tl) crystal is used as the detector. The background in the CsI(Tl) crystal is reduced by means of a brass shielding slab. The tritium target and separated isotope target foil lie in the plane of this slab while the CsI(Tl) crystal lies in the geometrical shadow of the slab, with its plane parallel to the surface of the slab (Fig.I.1)

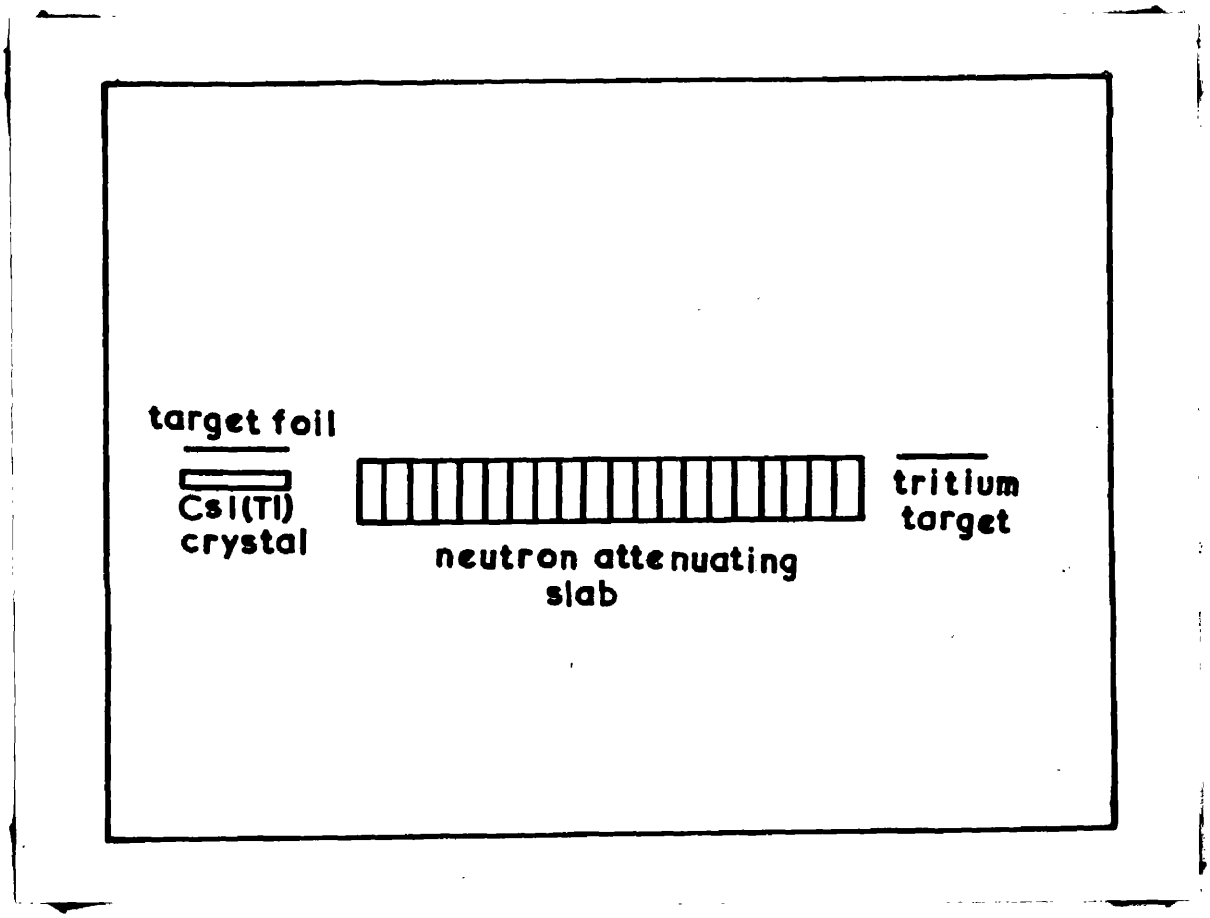


Figure I.1. Shielding arrangement used by Storey et al. (1960) in measuring the proton energy spectra from medium weight elements on bombardment with 14 MeV neutrons.

The solid angle is approximately π. The flux of neutrons at the crystal is then considerably attenuated relative to the flux at the target. Considering the extremely anisotropic angular distribution of elastically scattered fast neutrons, there may be an appreciable number of elastically scattered neutrons in the neighbourhood of the target foil. This will affect cross-section measurements to some extent, but not the energy resolution, since these neutrons scattered in the forward direction will have practically their original energy due to the small centre of mass effect involved. This method was designed to avoid contributions to the spectra from direct interactions and proved successful, good statistics being readily obtained. Again, however, it cannot be used to make measurements on the energy spectra below 4 MeV., because of the very intense β and γ ray backgrounds. As mentioned earlier, it is desirable to make such measurements. In this method no attempt is made to distinguish between different types of particle. It is assumed that because of the smaller cross sections, and in the case of alpha particles the smaller effective thickness of the target, contributions to the spectra from n-d, n-α etc. reactions can be ignored.

Many n-α, n-p and n-d cross sections have been measured using activation methods. A sample of the material is irradiated and the radio-activity produced is measured. Paul & Clarke (loc.cit.) and Coleman et al. (loc. cit.) have used this method fairly extensively. Where necessary radio-chemical techniques are employed.

I.4. The need for the present programme.

We can say that, at the time the author began his research, the difficulties encountered in experiments with fast neutrons had to a very large extent been surmounted, particularly in n-p experiments, by the techniques described in the previous section. Most of these are, of course, perfectly general and can be applied to experiments not involving neutrons. Measurements on the energy spectra of the alpha particles emitted in n-α reactions had not been made, however, mainly because of the very low cross sections for the reactions and the very thin target foils which must be used. These factors make it very difficult to obtain a suitable signal to background ratio. The proton background in the detector would be expected to mask the alpha particle spectra. The very short range of the alpha particle in nuclear emulsions inhibits the use of this technique. In addition, measurements using electronic techniques on the proton energy spectra emitted in n-p reactions had, as we have seen, been limited to energies greater than 4 MeV.

It was desirable that a method be found of making such measurements.

The generally accepted theory of such reactions at intermediate energies with medium weight elements is the statistical theory as described in Blatt & Weisskopf (1952) and attributed to Weisskopf (1937). The theory is based on the assumption that the nuclear reaction is separable into two distinct stages, the formation and decay of a compound nucleus, the decay being independent of the mode of formation and being determined principally by the availability of levels in the residual nucleus. It has become clear, however, from measurements of the energy spectra of the particles emitted in reactions such as (p,p'), (n,p), (α,p) etc. that this theory in its original form is inadequate to explain all of the experimental results.

This situation has led to other possible explanations of the mode of these reactions. It has been suggested that a direct interaction process takes place in which no compound nucleus is formed. This direct interaction may be a surface direct interaction (McManus & Sharp, 1952; Austern et al. 1953; 1958 Butler et al., 1958) or a volume direct interaction (Hayakawa et al., 1955; Brown & Muirhead, 1957). Some doubt has been cast on the validity of the volume direct interaction theories by the work of Elton and Gomes, 1957, and Glendenning, 1959.

Several authors, (Colli et al., 1957b; Thomson, 1956; Gugelot, 1954; Cohen, 1953), have suggested that Weisskopf's theory should perhaps be modified since the decay of the compound nucleus might be governed by selection rules which depend upon the energy of the emitted particle.

No model exists, however, which can explain all the observed experimental results and any further results which substantiate one or other of the proposed theories will be most useful.

In particular, measurements on n-α reactions would be most helpful since this reaction might be expected to proceed almost entirely by compound nucleus formation and the results obtained would be expected to agree with the predictions of the statistical theory. If this is not the case the implication would be that alpha particles exist inside the nucleus. Measurements of the energy spectra of protons of energy less than 4 MeV, emitted in n-p and n-np reactions could again be compared with statistical theory predictions and would be expected to be in fairly good agreement with these predictions.

To make such measurements, a new technique was required which could discriminate against electrons in the presence of protons, and protons in the presence of alpha particles. A method for distinguishing between particles had been

suggested by Wilkinson (1952). In this method a sandwich of two crystals, called a phoswich, is made. The luminescence (in one of the crystals) has a characteristic slow decay time while the luminescence in the other crystal has a characteristic fast decay time. Particles passing through both crystals produce a pulse with a decay time containing the characteristics of both crystals while particles stopping in or passing through a single crystal produce a pulse characteristic of that crystal. Electronic techniques are then used to distinguish between the different types of pulse. This technique has been used by Whetstone et al. (1958) as a photo-proton scintillation spectrometer. The phoswich, in this case, consists of NaI(Tl) and a plastic scintillator. The range of the NaI(Tl) crystal is exactly that of the highest energy proton to be detected. All protons therefore stop in the NaI(Tl) crystal, producing a pulse with the characteristic decay time of the luminescence in NaI(Tl). Electrons however pass through the NaI(Tl) crystal into the plastic and produce a pulse with a decay time containing the characteristics of both crystals. This results in a spike being produced in the pulses due to electrons. An electronic technique then distinguishes between proton and electron induced pulses. This technique did not seem particularly suited to neutron work however because of the high background which would be produced in

the phoswich. In the above form it is also unsuitable for distinguishing protons from alpha particles although, with modification, this could perhaps be accomplished.

Storey et al. (1958), working at Glasgow, found, shortly after the author began his research, that the decay time of the luminescence produced in a crystal of CsI(Tl) by an ionizing particle, is dependent on the density of ionization produced by the particle. The decay time of the luminescence is therefore different for alpha particles, protons and electrons. This was clearly an additional factor which might be developed to overcome the difficulties mentioned above and make it possible to extend the measurement of the energy spectra of protons emitted in n-p reactions to lower energies, and perhaps provide the first method of measuring the energy spectra of alpha particles emitted in n-α reactions. Alpha particles could be discriminated from protons and protons could be discriminated from low energy electrons, by the decay time of the luminescence they produce in CsI(Tl).

I.5. Present Programme.

The author therefore undertook the development of an electronic technique which would employ this difference in the decay time of the luminescence to discriminate one type of particle from another. We have seen that there was

a need for such a technique in low energy neutron physics. The technique would, of course, be perfectly general in application and could be applied equally well to γ-p measurements, for example, where the background of low energy electrons is again inhibiting. If successful the technique would be a useful and valuable instrumental addition to the commonly used techniques in low energy nuclear physics. Essentially the technique would consist of a single CsI(Tl) crystal capable of measuring the energy loss, $\frac{dE}{dx}$, and the energy, E, of a particle. It would constitute an advance in the technique of scintillation counting. The development of this technique was the main purpose of the work described in Part I of this thesis.

Part 1.

Particle Selection in Crystals of CsI(Tl).

Chapter II.

The work described in this chapter is the development of an electronic technique, suitable for use in low energy nuclear physics experiments, which uses the difference in the decay times of the luminescence produced in crystals of CsI(Tl) by electrons, protons and alpha particles to distinguish electrons from protons, electrons from alpha particles, and alpha particles from protons.

II.1. Experimental Technique.

II.1(a). 'Current' and Voltage' Pulses.

It will be advantageous to first consider the two types of pulse which can be obtained from a photo-electric multiplier tube, used in a scintillation counter. The first we shall call the 'voltage' pulse, the second we shall refer to as the 'current' pulse.

The 'voltage' pulse is obtained by arranging the time constant at a dynode of the photomultiplier to be very long in comparison with the decay time of the luminescence produced in the luminescent material. The rise time of this pulse is then governed by the decay time of the luminescence and will be different for pulses produced by

different types of particle. The 'voltage' pulse decays with the electronic time constant at the dynode.

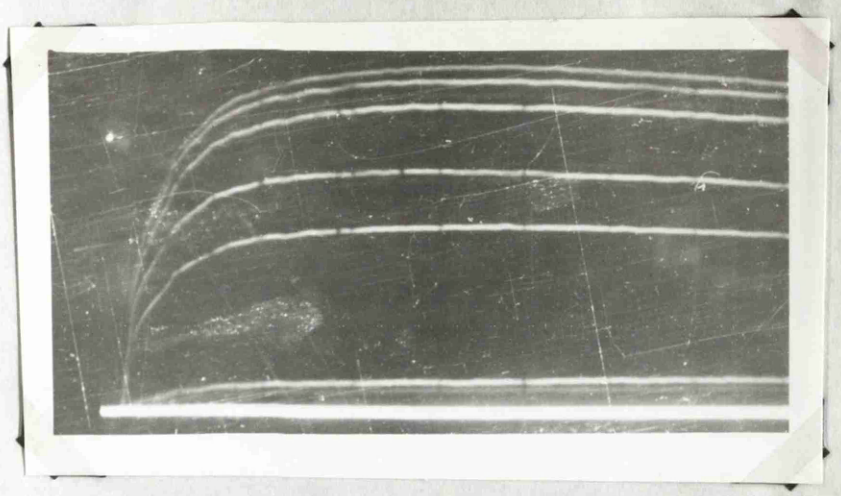
The 'current' pulse is obtained by arranging the time constant at a dynode of the photomultiplier to be very short in comparison with the decay time of the luminescence. The rise time of this pulse is governed by the transit time of the photomultiplier and the rise time of the luminescence. The decay time of this pulse is equal to the decay time of the luminescence and is therefore different for pulses produced by different particles (Storey et al. loc. cit.).

The 'voltage' pulse is in fact the integrated 'current' pulse.

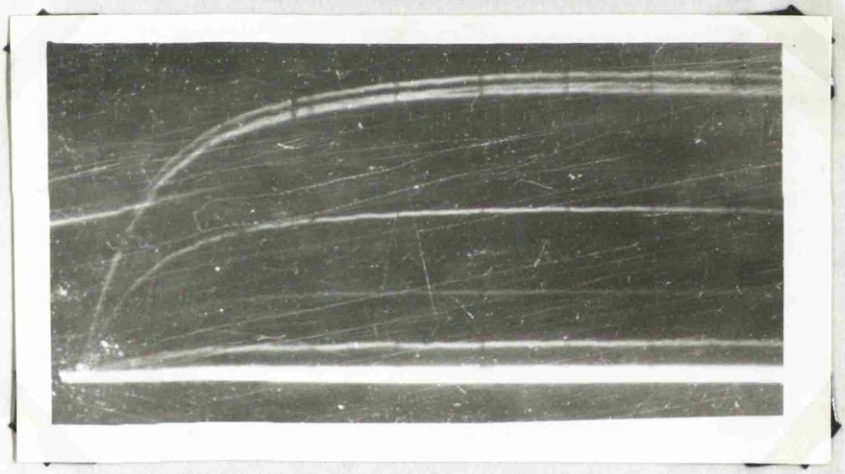
Both the 'current' and 'voltage' pulse are used in the course of the work described in this thesis. Photographs of these pulses produced by alpha particles and electrons from an RCA 6810 photomultiplier are shown in figure II.1a,b using CsI(Tl) as the luminescent material. The difference in the decay times of the luminescence produced by alpha particles and electrons is readily seen.

II.1(b). Alpha particle - electron discrimination.

The decay of the luminescence produced in standard crystals of CsI(Tl) by ionizing particles was found by

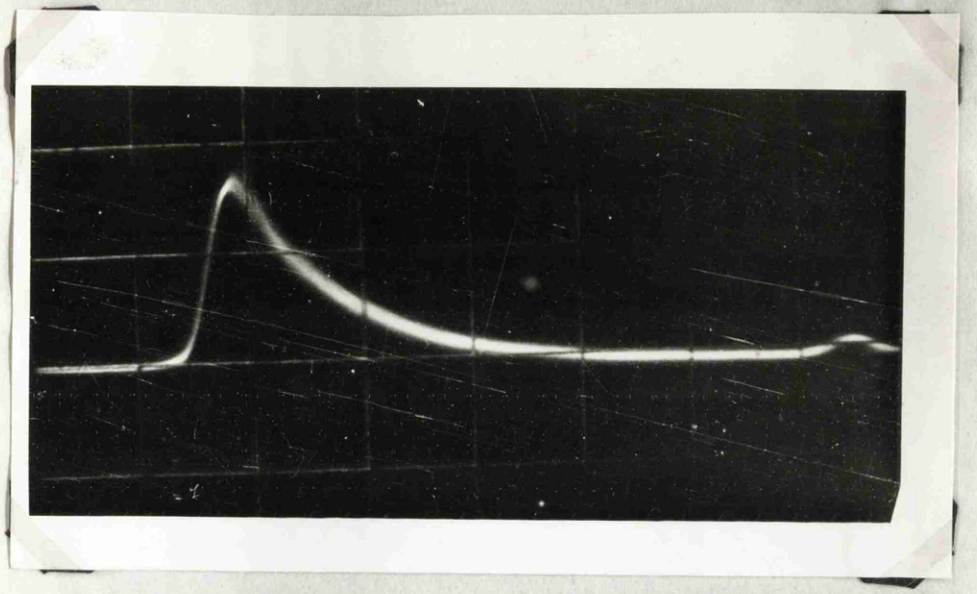


Voltage pulse produced by alpha particles.

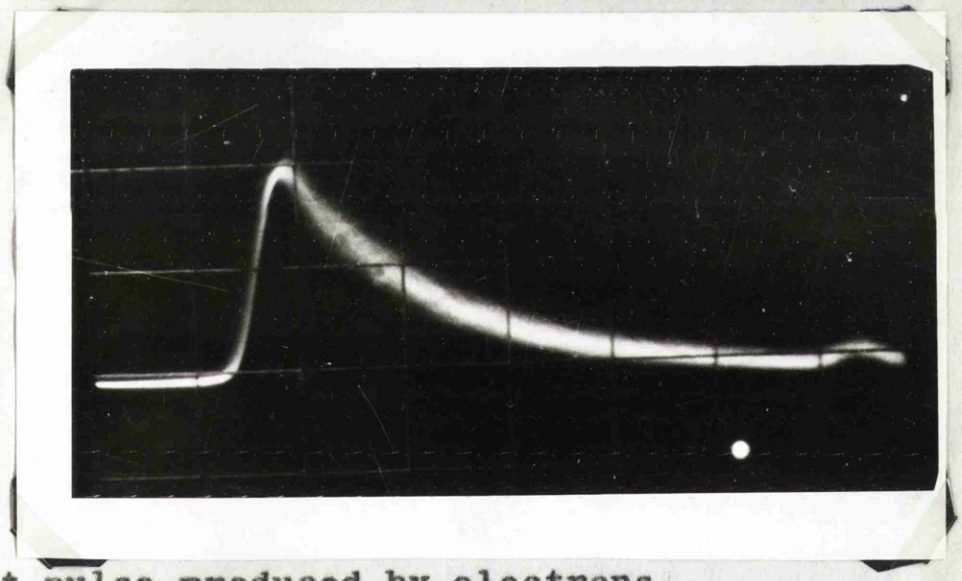


Voltage pulse produced by electrons.

Figure II.1(a). Photographs of the 'voltage' pulses produced by electrons and alpha particles from an R.C.A. 6810 photomultiplier, using CsI(Tl) as the luminescent material. The time scale on the oscilloscope was 2 μs/cm.



Current pulse produced by alpha particles.



Current pulse produced by electrons.

Figure II.1(b). Photographs of the 'current' pulses produced by electrons and alpha particles from an R.C.A.6810 photomultiplier using CsI(Tl) as the luminescent material. The time scale on the oscilloscope was 0·65 μs/cm.

Storey et al. (loc. cit.) to consist of two exponential components τ_1 and τ_2 .

The values of the short and long components, τ_1 and τ_2 respectively, of the decay time of the luminescence produced by alpha particles and electrons (~~under~~ molar concentration of thallium 0.1%) are limiting values of: $\tau_{1\alpha}$ = 0.42 μ.sec.; τ_{1e} = 0.7 μ.sec.; $\tau_{2\alpha,e}$ = 7 μs. The subscripts are used to denote the type of particle producing the luminescence. This difference in the values of $\tau_{1\alpha}$ and τ_{1e} has been used in the technique described below to discriminate alpha particles from electrons. A block diagram of the apparatus is shown in figure II.2. Voltage pulses from the photomultiplier R.C.A. 6810A are recorded in the usual way by the pulse height analyser (Hutchinson-Scarrott type) when a gate is supplied by a circuit which effectively inspects the decay time of the 'current' pulse from the photomultiplier. The circuit is arranged to produce no gate when the original current pulse was due to an electron in the CsI(Tl) crystal. This is done by clipping the current pulse at the peak height to obtain a narrow pulse of the opposite polarity which is used to charge up a condenser through two crystal diodes. The value of a leakage resister in parallel with the condenser can be varied (or tuned) to make the decay time of the pulse across the condenser coincide with the

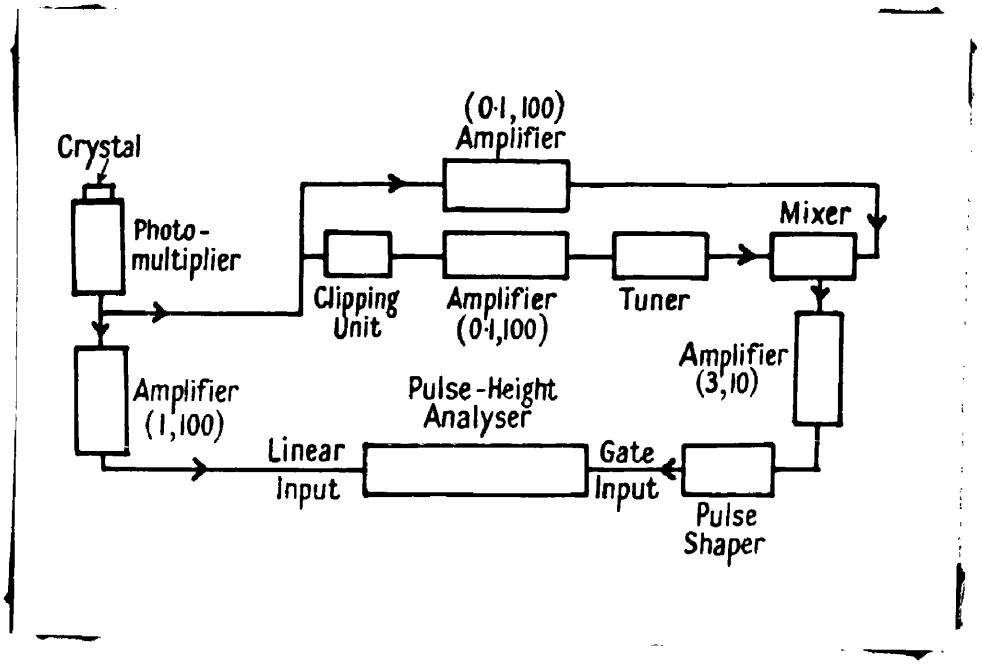


Figure II.2. Block Diagram of the apparatus. The numbers shown in brackets are the differentiation and integration time constants in the amplifiers. The integration time constant is given first.

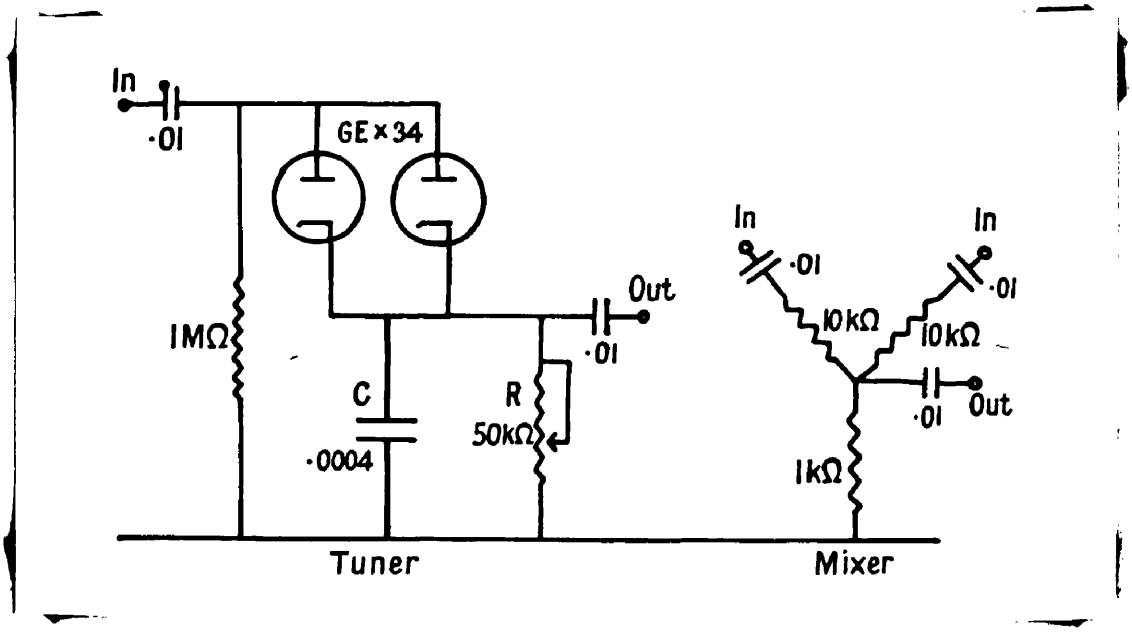


Figure II.3. Circuits for the 'tuner' and 'mixer' in figure II.2.

value of τ_{1e} . When this pulse is mixed with the original current pulse the sum of the two is very small for electrons and sufficiently large for alpha particles to overcome the bias in a discriminator on the input of a pulse shaper which supplies a suitable gating pulse to the pulse height analyser. The circuits for the 'tuner' and 'mixer' in figure II.2 are very simple but are given for convenience in figure II.3. The amplifiers used are I.D.L. type No.652. The clipping unit is a standard circuit using three CV. 138 valves. The pulse is clipped at the anode of the first valve, inverted in the second valve and fed to the next stage of the circuit via the third valve, which acts as a cathode follower. The pulse shaper which supplies the gate for the kicksorter consists simply of a flip flop designed to give a 50 volt negative pulse, rising to 20 volts in 0.1 μ.s., suitable for gating the kicksorter.

The operation of the circuit is illustrated by the pulse height distributions in figures II.4 and 5 which were obtained using alpha particles from Po^{210} and gamma rays from ThC^{11} (E_γ = 2.6 and 0.5 MeV). These clearly demonstrate the ability of the circuit to detect only alpha particles in the presence of strong electron backgrounds. Figure

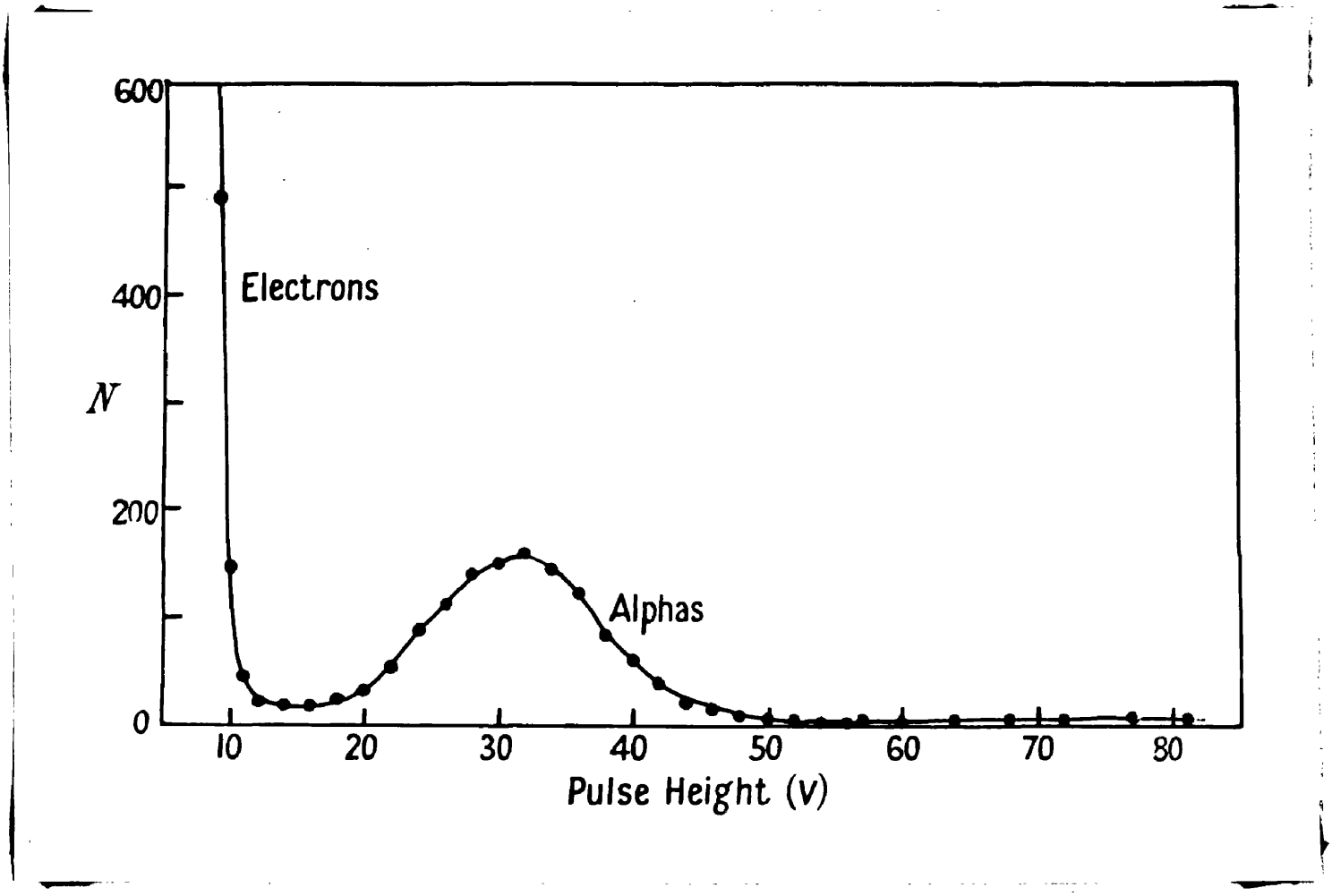


Figure II.4. Pulse height distribution of pulses from the 'mixer' due to 2.5 MeV alpha particles and 0.58 MeV and 2.6 MeV gamma rays.

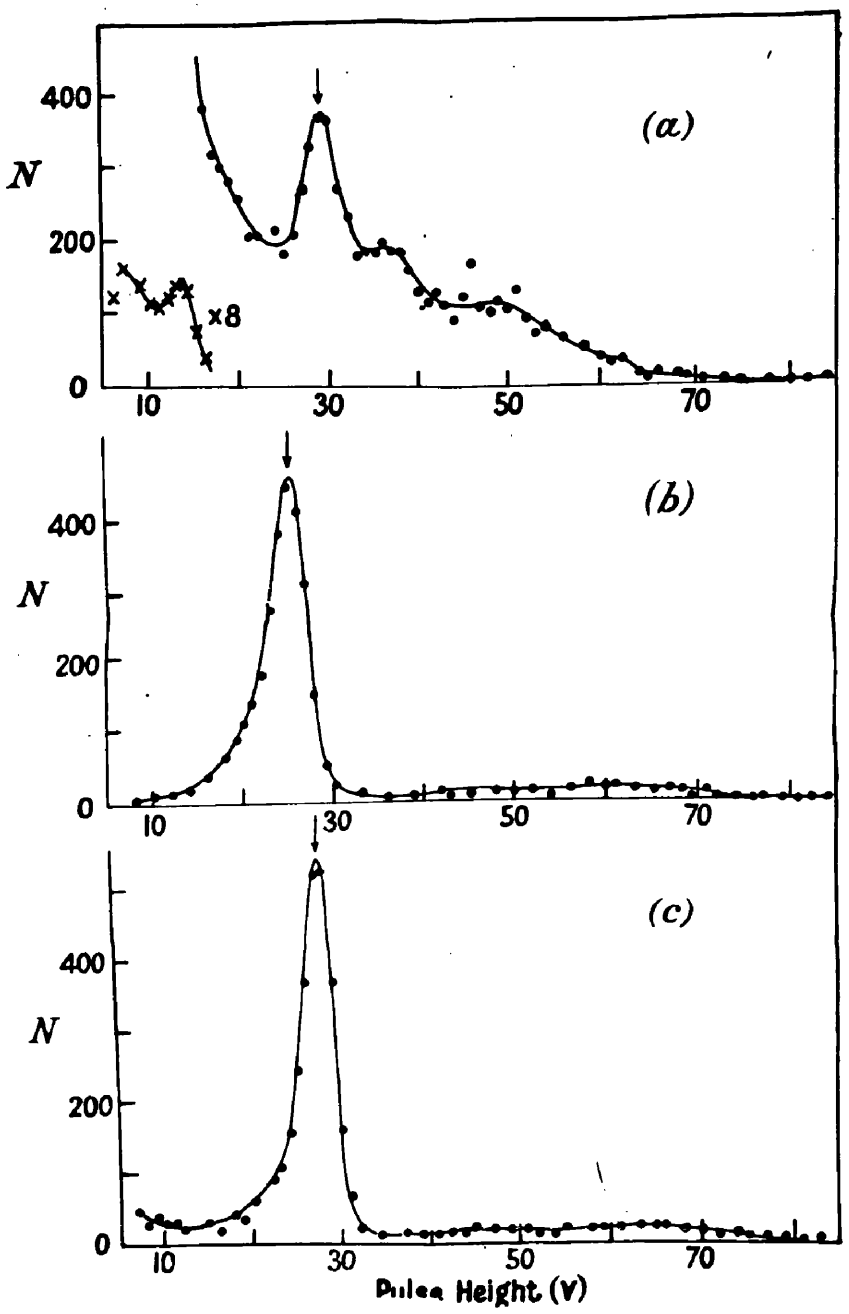


Figure II.5. Pulse height distributions of the 'voltage' pulses from the photomultiplier. In (a) and (b) the CsI(Tl) crystal was exposed to alpha particles and gamma rays for the same time and under the same conditions in which the distribution shown in figure II.4 was obtained. In (a) all pulses were recorded by the pulse height analyser. In (b) the gating pulse was used to select only pulses due to alpha particles. In (c) the gamma ray source was removed and all pulses recorded for the same time as in (a) and (b). The peak due to 2.5 MeV alpha particles is indicated by an arrow, pulses above this peak in (b) and (c) are due to higher energy alpha particles.

II.4 is the pulse height distribution of pulses from the 'mixer' when the CsI(Tl) crystal was exposed to 2.5 MeV alpha particles and gamma rays. It can be seen that the bias on the discriminator following the 'mixer' can be set above the height of pulses due to electrons. Statistical fluctuations in the peak heights of the original current pulses are the main cause of the relatively large width of the peak due to alpha particles and the finite size of pulses due to electrons. The latter sets an upper limit to the electron energy if pulses due to electrons are to be avoided completely. A further cause of the finite size of pulses due to electrons is the nature of the decay of the luminescence produced by electrons in the CsI(Tl) crystal. This decay is not purely exponential in form but, as mentioned earlier, is composed of two decay time components, τ_{1e} and τ_{2e} . The 'tuned' pulse is at the best an approximation to this complex decay being purely exponential in form. This causes a mismatch in the two pulses arriving at the mixer. For this reason it is perhaps surprising that the method works at all.

The pulse height distributions shown in figure II.5 are for the voltage pulses from the photomultiplier. Figure II.5(a) shows the usual distribution obtained by allowing the pulse height analyser to record all pulses.

Figure II.5(b) shows the effect of the gating pulse in selecting only pulses due to alpha particles. The position of the gamma ray source and the polonium source were kept fixed in obtaining the distributions in figures 4, 5a and b, and the counting time in each case was two minutes. The gamma ray source was removed to obtain the distribution in figure II.5(c) which was measured by allowing the pulse analyser to record all pulses again for two minutes. The points in figures II.5(b) and (c) above the peak for 2.5 MeV alpha particles are due to alpha particles from polonium on the thin foil reflector above the CsI(Tl) crystal. The success with which alpha particles can be detected in the presence of strong electron backgrounds is readily seen. A comparison of figures II.5(b) and (c) shows that the suppression of the electron background has not interfered with the alpha particle spectrum except that the coincidence technique has produced a very slight decrease in the resolution. By 'tuning' to the decay time component $\tau_{1\alpha}$ instead of τ_{1e} , we could discriminate against alpha particles. We are unlikely, however, to wish to do this in any nuclear experiment.

Further tests were now made to see if the technique could be used to distinguish protons from electrons, and protons from alpha particles.

II.1(c). Proton-Electron Discrimination.

Before the technique could be said to be satisfactory three tests were necessary.

For protons the value of the short component of the decay of the luminescence produced by protons, τ_{1p}, varies with the energy of the proton according to the density of ionization produced in the CsI(Tl) crystal. For high energy protons ($\gtrsim$ 15 MeV) τ_{1p} would be very similar to τ_{1e}. In an experiment using the technique described above to eliminate electron backgrounds it might therefore be possible to detect only protons of less than a certain maximum energy since high energy protons might be removed as electrons. Tests were made using protons of energy 14.8 MeV obtained from the $^{3}He(d,p)^{4}He$ reaction (Q = 17.8 MeV). It was found that these protons could easily be discriminated from the electrons produced in the crystal by gamma rays from the ThC^{11} source and were detected with 100% efficiency. The pulse they produced from the 'mixer' was in fact of the same magnitude as that produced by an alpha particle of energy 2 MeV.

When the gains of the R.C.A. 6810A photomultiplier and the I.D.L. amplifiers used in the circuit are adjusted so that the pulses produced from the photomultiplier by protons of energy $\leq$ 15 MeV are not saturated, it is clear

that protons of less than a certain minimum energy will not be discriminated from electrons with 100% efficiency. The statistical fluctuations on the pulses produced by protons of energy $\leq$ this minimum energy will become very serious and the detection efficiency will gradually decrease as the proton energy decreases. Figures II.6(a) and (b) show the results of tests to find this minimum energy. These figures show that protons of energy 1.5 MeV, obtained from the reaction $^{2}H(^{2}H,p)^{3}H$ (Q = +4.04 MeV), can be readily discriminated from the intense electron backgrounds produced in the CsI(Tl) crystal by the gamma rays from the ThC^{11} source. In obtaining the spectra shown in figures II.6(a) and (b), the deuteron beam in the H.T. set was not monitored absolutely, a steady beam was maintained for five minutes. The spectra shown are in fact the average of six single runs in each case. It was found that the variation in the single spectrum was less than 4%. This variation was greater than the variation between the single and coincidence spectra and it seemed reasonable to assume that the proton detection efficiency was 100%. Protons of energy 1/2 MeV were in fact detected with only 90% efficiency, due to the statistical fluctuations on the pulses produced from the photomultiplier. This results in a spread in the size of the pulses they produce

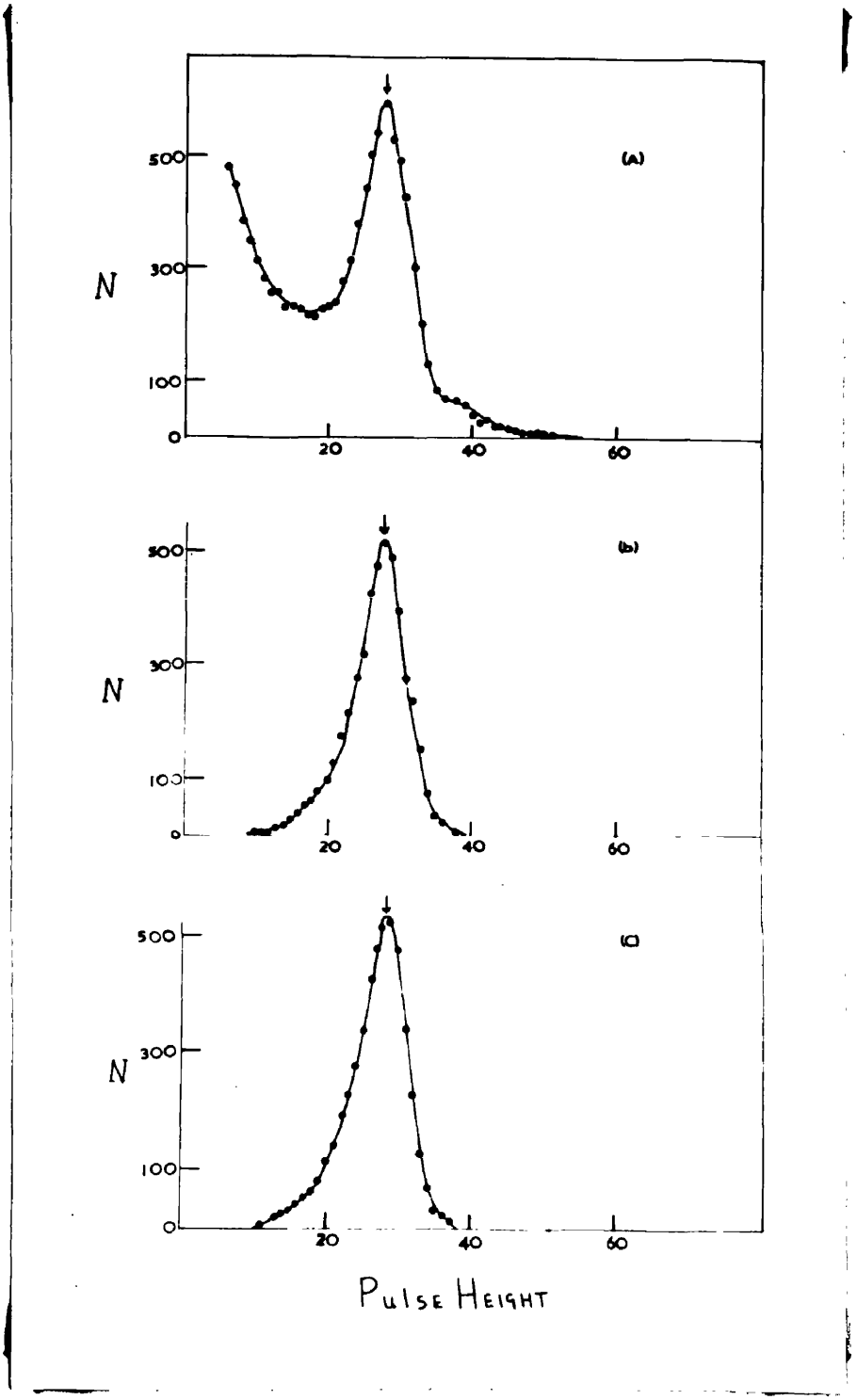


Figure II.6. Pulse height distributions of the 'voltage' pulses from the photomultiplier. In (a) all pulses were recorded by the pulse height analyser. In (b) the gating pulse was used to select only pulses due to protons. The peak due to 1.5 MeV protons is denoted by an arrow. The gamma rays were obtained from a ThC'' source.

from the mixer, as illustrated in figure II.4 for 2.5 MeV alpha particles. The pulses from the mixer do not, as a result, overcome the bias level in the discriminator following the mixer, set to reject electron pulses from the mixer, with 100% efficiency and the detection efficiency correspondingly decreases. Protons of this energy can be detected with 100% efficiency by increasing the photo-multiplier gain bit then the pulses produced by high energy protons are saturated.

These tests showed that protons of energy 1-15 MeV could be simultaneously discriminated, using the above technique, from the electron backgrounds produced in a CsI(Tl) crystal by gamma rays of energy 1-3 MeV.

The third, and perhaps most important test, was now made. A study of the circuitry shows that the magnitude of the pulse produced from the 'mixer' is proportional to the energy of the exciting particle. It is true that when we are 'tuned' to the value γ_e a null should, theoretically be produced from the 'mixer' by all energies of electrons. We have seen, however, for the reasons given in section II.1(b), that this is not the case. The electron produced pulses from the 'mixer' are in fact of finite size and are proportional to the initial energy of the electrons. Thus a bias setting in the discriminator following the 'mixer',

set to reject pulses produced by 3 MeV gamma rays, might not be sufficient to reject pulses produced by 6 MeV gamma rays. The technique would as a result be limited in energy range. In fact tests indicated, that, in its above form, the technique was indeed limited in energy range. It was found, for example, that if the bias setting in the discriminator following the mixer was set to reject pulses produced by 6 MeV gamma rays with 100% efficiency, then the lowest energy protons detectable with 100% efficiency were of energy 4 MeV. Protons of smaller energy did not produce a pulse from the mixer of sufficient magnitude to overcome this bias setting and the detection efficiency gradually fell off. In a similar manner the detection efficiency for higher energy gamma-rays gradually increased.

Summarising, one can say that protons of energy up to at least 14 MeV can be discriminated from electrons using the difference in the decay times of the luminescence they produce in CsI(Tl). The technique described above for doing this, however, is in its above form very limited in energy range ($\varkappa$ 2 MeV).

II.1(d). <u>Alpha Particle-Proton Discrimination</u>.

The tests described in section II.1(C) had to be repeated for this case.

In this case, however, γ_{lp} for low energy protons is

very similar to $\gamma_{1\alpha}$ and we must find the minimum energy which protons can have and still be discriminated from alpha particles with 100% efficiency. Tests showed that this minimum energy was 3.5 MeV. For protons of lower energy γ_{1p} is almost identical to $\gamma_{1\alpha}$, and statistical fluctuations make it impossible to discriminate these low energy protons from alpha particles with 100% efficiency.

This minimum energy of 3.5 MeV corresponds to an alpha particle energy of 5 MeV (Fulmer and Cohen, 1958) and as a result the use of a technique of the type described above in discriminating alpha particles from protons is limited to alpha particles of energy greater than 5 MeV.

The energy range of the technique, in its above form, in discriminating between alpha particles and protons is illustrated in figures II.7(a) and (b). In this case we were tuned to $\gamma_{1\alpha}$. Figure II.7(a) shows the spectra obtained when a lithium target was bombarded by deuterons, allowing the pulse analyser to record all pulses. In this spectrum the proton groups at 4.1 and 4.6 MeV correspond to transitions to the ground state and first excited state of ^{7}Li from the ground state of ^{6}Li, by the reaction ^{6}Li(d,p)^{7}Li (Q = 15.02 MeV). The alpha particle group at 8 MeV corresponds to transitions to the ground state of ^{4}He by the reaction ^{6}Li(d,α)^{4}He (Q = 22.375 MeV). The low energy

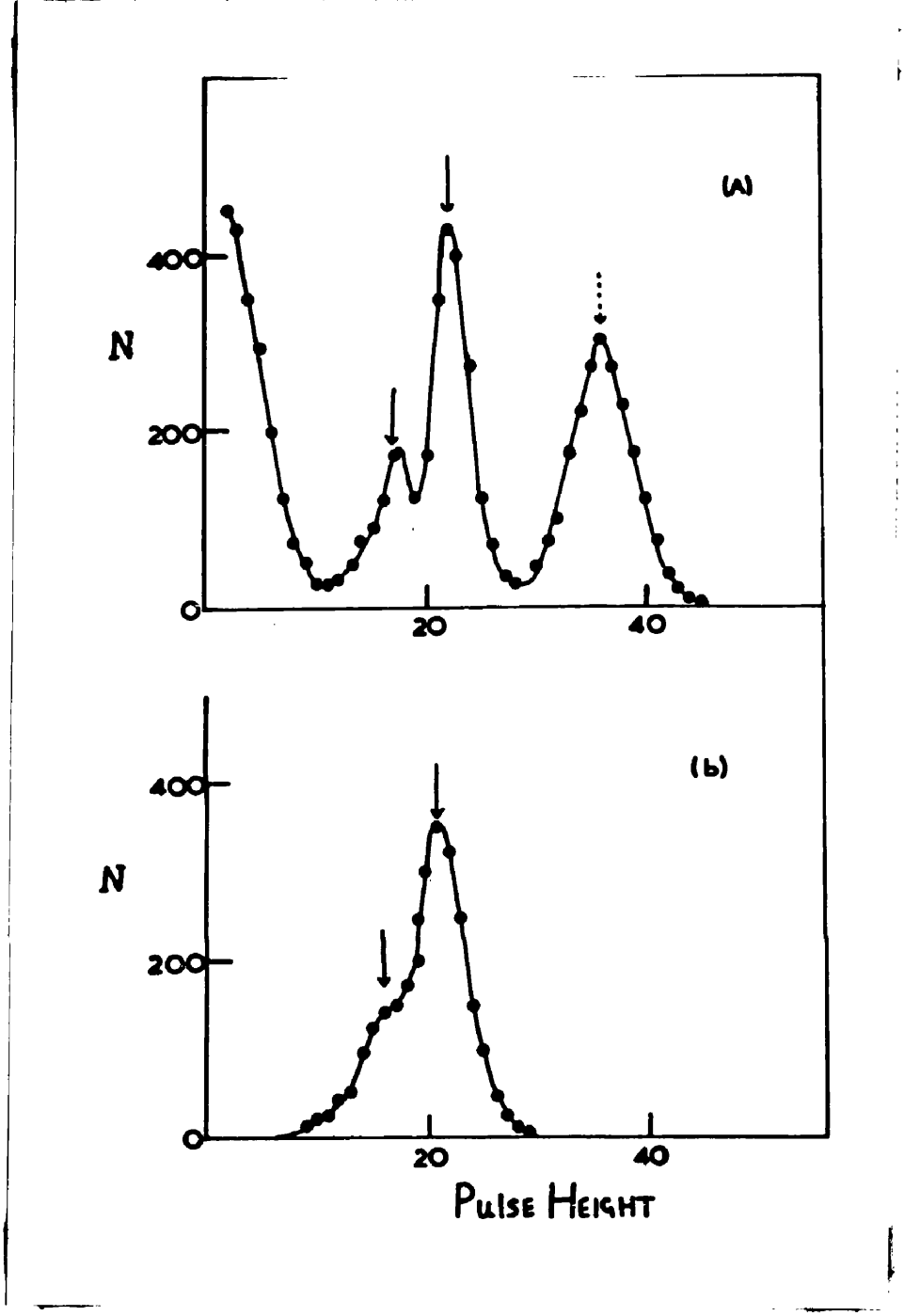


Figure II.7. Pulse height distribution of voltage pulses from the photomultiplier. In (a) all pulses were recorded by the pulse height analyser. In (b) the gating pulse was used to select only pulses due to protons. The peaks due to protons of 4.1 MeV and 4.6 MeV are denoted by solid arrows. The peak due to 8 MeV alpha particles is denoted by the dotted arrow. The low energy pulses in (a) are due to electrons. These are eliminated in (b) because they do not produce a pulse of sufficient magnitude from the mixer to overcome the bias in a discriminator set to reject the high energy alpha particles.

pulses are due o low energy electrons and gamma rays. Figure II.7(b) clearly shows the alpha particle group at 8 MeV being eliminated leaving the proton groups at 4.1 and 4.6 MeV. The resolution has been slightly impaired by the coincidence technique. The low energy pulses due to electrons and gamma rays were also eliminated. In this case this was due to their not producing a pulse of sufficient magnitude from the 'mixer' to overcome the bias in the discriminator following the 'mixer', set to reject the high energy alpha particles. This again demonstrates the dependence on energy of the magnitude of the pulse from the 'mixer'. In a similar fashion the detection efficiency for protons of energy less than 2.5 MeV gradually decreases. The detection efficiency gradually increases for alpha particles with energy greater than those in the peak of figure II.7(a).

The spectra shown in figures II.7(a) and (b) are again in each case the average of six five minute runs. Again the variation in the number of protons in the peaks in the single spectra obtained, was greater than the variation between the single and coincidence spectra and it again seemed reasonable to assume 100% detection efficiency for the protons.

Summarising one can say that alpha particles of energy greater than 5 MeV can be discriminated from protons by the difference in the decay times of the luminescence they produce in CsI(Tl). The range of the technique described above is limited to about 4 MeV.

A final test of the technique, in its above form, was then made in which alpha particle, proton and electron produced pulses of equal height were passed through the apparatus. The circuit was found to separate these in the expected manner. No pulse was produced from the 'mixer' for electrons. The proton pulse from the 'mixer' varied in magnitude according to the energy of the proton, but was always less than the pulse produced from the 'mixer' by alpha particles.

II.2. Possible Improvements of the Technique.

The results presented in the previous sections were encouraging. Even with its limited energy range, the technique in its above form would have been of some use in nuclear physics. It could have been used, for example, to permit the measurement of the angular distribution of alpha particle groups, corresponding to transitions in nuclei, which would otherwise be masked by proton groups. Again in an n-p experiment at 14 MeV, in which the measurement of the proton spectra is inhibited below 4 MeV by the low energy electron background, the technique

could have been used to extend the measurements to 2 MeV. The limited energy range was, however, undesirable and for this reason possible improvements in the technique were considered.

The technique could possibly have been improved by making use of the fact that the greater in magnitude the pulse from the 'mixer', the greater, also, is the magnitude of the original voltage pulse from the photomultiplier. Feeding the pulse from the mixer to the grid of a valve and the original voltage pulse, arranged to be of opposite polarity, to the screen of this valve would produce a pulse from the anode of this valve whose magnitude is independent of the energy of a particle but is dependent solely on particle type. This pulse could then be used to provide a gate for the pulse height analyser in the usual way. This would overcome the previous difficulty of limited energy range.

A second possibility was the following. If, for example, discrimination was required between protons and electrons, then instead of 'tuning' precisely to γ_{1e} so as to produce a null from the 'mixer' for electrons, we could tune to a value between γ_{1e} and γ_{1p} so that protons and electrons would produce pulses of opposite polarity from the 'mixer' as illustrated in figure II.8.

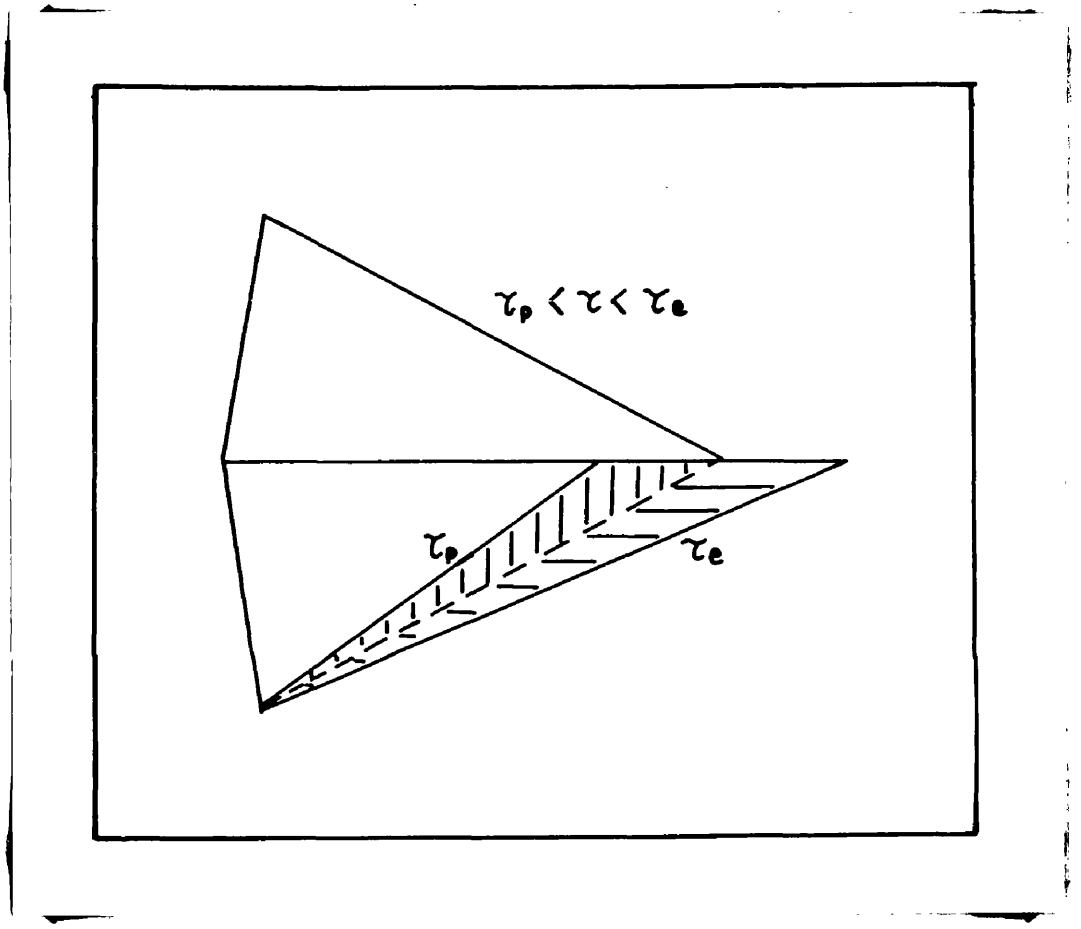


Figure II.8. By tuning to a value of τ between τ_{ie} and τ_{ip}, pulses are obtained from the 'mixer' which are proportional to the shaded areas. The pulses from the 'mixer' are positive for protons and negative for electrons.

In this way, the major difficulty of the 'null' method of tuning would be overcome. The polarity of the pulse from the 'mixer' would depend on the decay time of the luminescence produced by a particle and its magnitude would then be unimportant.

Finally, changing the thallium concentration in the CsI(Tl) crystal, or altering its temperature, might have resulted in a greater difference in the decay times of the luminescence produced by different types of particle, and pulse shape discrimination would clearly have been much easier. Again, it was possible that another crystal in which the decay time was more dependent on the ionization density could have been found. (Experiments on the luminescence of alkali halide crystals were in fact made to investigate these possibilities. A description of these experiments forms the basis of part II of this thesis entitled:- Associated Experiments on the Luminescence of Alkali Halides. It suffices, at present, to remark that, in fact, no improvement can be made in the technique in any of these ways).

II.3. Final Form of the Technique.

It is found that the second method for improving the technique, discussed in the previous section, proves very successful. The circuitry is identical to that in figure II.2. In discriminating between protons and electrons, the RC time constant in the 'tuner' is arranged to be intermediary in value between τ_{1p} and τ_{1e} . Electrons, after amplification, then produce positive pulses from the mixer while protons produce negative pulses. Photographs of the pulses produced by electrons from Th C^{11} and 2.5 MeV protons are shown in figure II.9. This positive pulse, produced from the 'mixer' by electrons, is then used to produce an anti-coincidence in a proportional gate circuit and in this way electron produced pulses are not recorded on the pulse height analyser. In a similar way discrimination is possible between protons and alpha particles, and alpha particles and electrons. The proportional gate circuit is shown in appendix I.

Used in this manner the energy range of the technique is very considerably increased.

Tests showed that in discriminating between protons and electrons, we could readily discriminate against all electrons with energies greater than 1 MeV, at the same time detecting all protons with energies between 1-15 MeV. The upper limit on the proton energy is due to the similarity in the decay of the luminescence produced by electrons and high energy protons. The lower limit on the

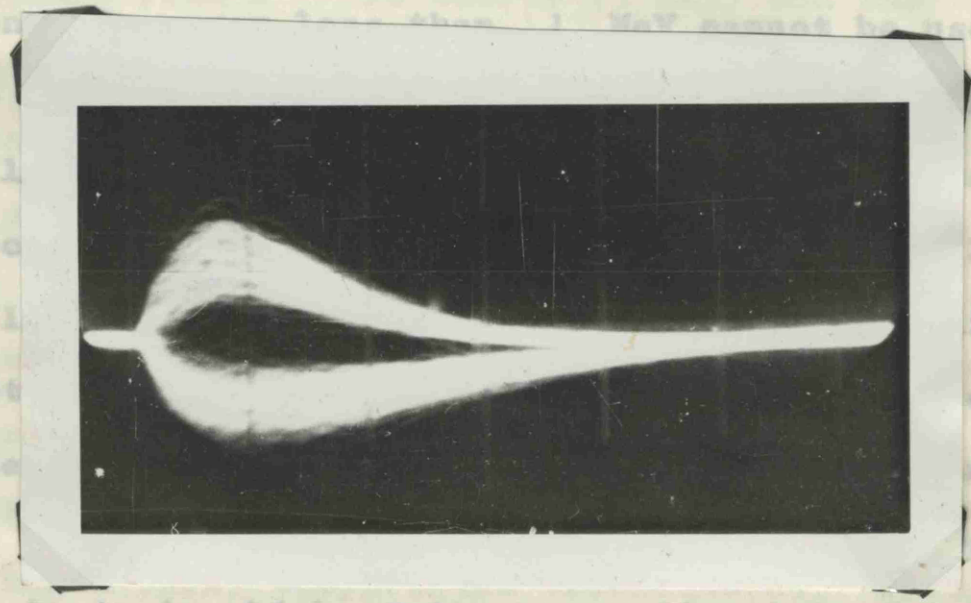


Figure II.9. Photograph of the pulses produced from the 'mixer' by 2.5 MeV protons and electrons from the ThC^{11} source. In this photograph the electron pulses are positive, the pulse from the 'mixer' having been inverted and amplified in an I.D.L. amplifier. The time scale on the oscilloscope was 2 μs/cm.

electron energy is a result of the fact that when the gains of the photomultiplier and amplifier are set so that pulses produced by protons of energy 15 MeV are not saturated, then the pulse produced from the 'mixer' by electrons of energy less than 1 MeV cannot be used to produce an anticoincidence in the gate circuit. The original voltage pulses produced by these electrons are then recorded on the pulse height analyser. This lower energy limit can be decreased by increasing the gain of the photomultiplier but only at the expense of rejecting high energy protons. The energy range of the technique is thus 1-15 MeV, more than sufficient for our purpose.

In tests in which protons were discriminated from alpha particles, it was found that we could readily discriminate against all protons with energies between $3\frac{1}{2}$-15 MeV at the same time detecting all alpha particles with energies in this range. At 3 MeV approximately 10% of the protons are detected as alpha particles. Because of the different response of CsI(Tl) to alpha particles and protons, this proton energy range corresponds to an alpha particle energy range of 5-16.5 MeV. The lower limit in the proton energy is, in this case, due to the similarity in the decay time of the luminescence produced by protons of less than 3.5 MeV and alpha particles.

The protons used in these tests were obtained from the $^{3}He(d,p)^{4}He$ and $^{2}H(^{2}H_{1},p)^{3}H$ reactions. The electrons were produced in the CsI(Tl) crystal by the gamma rays of energy 2.6 MeV from a ThC^{11} source. Alpha particles were obtained from a ^{210}Po source (5 MeV) and the $^{6}Li(d,\alpha)^{4}He$ reaction (8 MeV). High energy electrons and alpha particles were simulated by using the ^{210}Po and ThC^{11} sources and increasing the gain of the photomultiplier till the pulse heights they produced were the same as would have been produced by 15 MeV alpha particles and electrons respectively.

II.4. Discussion.

We now briefly compare the technique described above with similar techniques which have been developed in parallel with the author's research.

In CsI(Tl) the ratio of $\frac{\text{peak light intensity}}{\text{total light output}}$ is different for alpha particles, protons and electrons. R.B. Owen (1959) presents a circuit which uses this fact to discriminate between them. Owen does not examine the energy range over which the device operates but Dosch et al. (1960) have developed this circuit for use in a γ-p experiment at 35 MeV. Their results indicate that discrimination between protons and electrons is possible between at least $2\frac{1}{2}$-12 MeV. They hope to use it for

discriminating between alpha particles and protons in γ-α reactions, also at 35 MeV. The circuit appears to operate just as successfully as the one described above.

Becker (1960) discusses the use of a circuit developed by Biggerstaff & Becker (1959) for discriminating alpha particles from protons. This technique, however, has a very limited energy range.

Hrehuss (1960) describes a method of particle discrimination which uses the fact that, in the emission spectra of CsI(Tl) more shorter wavelength photons are produced by electrons than by particles producing greater ionization densities. Two photomultipliers view a single CsI(Tl) crystal. Between the crystal and the photomultipliers are placed filters which pass wavelengths $\lambda >$ and $<$ 500m μ respectively. From the multiplier with the filter passing wavelengths $<$ 500m μ a pulse is obtained which differs for different particles. From the other photomultiplier a pulse is obtained which is identical for all particles. These pulses are then used to discriminate between different types of particle. The lowest energy of alpha particle which can be discriminated from electrons is of energy 1.7 MeV, due to statistical fluctuations on the pulses. The method does not appear to be as successful as any of the other methods described.

Finally Owen (1959) and Brooks (1959) have developed a similar technique to discriminate neutrons from Y-rays. This depends on the difference in the luminescence produced in organic phosphors by protons and electrons. The neutron produces a ' knock on' proton in the organic phosphor which is then distinguished from the electrons produced by the Y-rays by the difference in the detailed composition of the decay of the luminescence they produce.

All of these particle discrimination techniques using CsI(Tl) as the phosphor suffer from two limitations:-
(a) Since the decay time of the luminescence produced by protons varies with energy, there is a lower limit to the energy of protons which can be discriminated from alpha particles and an upper limit to the energy of protons which can be discriminated from electrons.
(b) Because of the lengths of the decay times of the luminescence in CsI(Tl), the analysis time involved in all such techniques is of the order of a few microseconds. This might result in pulse pile up for very high counting rates.

It is fair to say that the use of pulse shape analysis discrimination constitutes an important advance in the

technique of scintillation counting and constitutes a useful instrumental addition to the techniques of low energy nuclear physics. The work presented in this chapter was the first technique of this nature to be published. It is also fair to say that the purpose of the work in this part of the thesis had been accomplished. The development of an electronic method for discrimination between different types of particles had been achieved.

Part I.

The Use of Pulse Shape Analysis Discrimination in Reducing the Backgrounds Produced in a CsI(Tl) Crystal on Irradiation with Neutrons.

Chapter III.

In this chapter, the use of the pulse shape discrimination technique is illustrated in reducing the unwanted, but unavoidable, backgrounds produced in a CsI(Tl) crystal on irradiation with neutrons. The technique is perfectly general in application in low energy nuclear physics but since we are primarily interested in its applications in neutron physics at 14 MeV, its use can best be illustrated in this manner.

III.1. The Backgrounds Produced in a CsI(Tl) Crystal on Irradiation with Neutrons.

We must first consider the nature of the backgrounds which are produced when CsI(Tl) is irradiated with neutrons. These will form the backgrounds in experiments of the type n-α, n-p etc. in which a CsI(Tl) crystal is used as the detector, and the reductions in these backgrounds produced by the use of the pulse shape analysis technique will be a measure of its use in neutron physics.

We expect the main contribution to the background produced to be protons from the reactions $^{133}Cs(n\ p)^{133}Xe$, and $^{127}I(n\ p)^{127}Te$. Alpha particle emission is inhibited

by the very large Coulomb barrier and the cross sections for the reactions $^{133}Cs(n,\alpha)^{130}I$ and $^{127}I(n,\alpha)^{124}Sb$ are as a result considerably smaller than the n-p cross sections. Deuteron emission would be expected to show itself in the form of structure in the spectra corresponding to transitions to single levels in the residual nucleus. At low energies we expect the background to consist almost entirely of low energy β and γ-rays.

In the following sections the results of measurements on these backgrounds are presented.

III.1(a). Experimental Arrangement.

The CsI(Tl) crystal (⅝1"x 0.4 mm) was mounted quite rigidly with 0.005" platinum wire in a brass chamber at a distance of 1.5" from the glass cap which formed the bottom of the chamber. The inside of the chamber was lined with silvered lead to act as a light reflector and at the same time to reduce the background in the CsI(Tl) crystal from external sources. A very thin aluminium foil (0.00003") was placed behind the crystal to act as a light reflector. The chamber was evacuated to remove any background due to n-p or n-d events in nitrogen in the air. A mylar window (1.28 mg/cm^2) allowed calibration protons to enter the crystal. This window was blocked off by a lead cap during irradiation with the neutrons. Optical contact was made between the glass cap of the chamber and an RCA 6810

photomultiplier tube with silicone grease. The photomultiplier and chamber were then assembled on a stand which permitted the crystal to be rotated about the neutron source. The angle between the incident deuteron beam and the crystal was very precisely known and hence the neutron energy. Measurements were made at 13.5, 14.1 and 15.1 MeV. The pulses from the photomultiplier were amplified using an I.D.L. amplifier and the pulse height distribution measured on a 100 channel pulse height analyser. Six runs were made at 13.5 MeV, five at 14.1 MeV and four at 15.1 MeV. The results of each run were first plotted separately to check for consistency, then averaged. The neutron monitor shown in figure III.1 was designed by Dr. R.S. Storey in this laboratory, to correspond to the accurate computations of Bame et al. (1957). A typical pulse height distribution for 14 MeV neutrons is shown in figure III.2, and it can be seen that the peak due to "knock on" protons from the polythene radiator gives this type of monitor a high stability. Here again the air was removed to avoid a small background due to n-p and n-d events in nitrogen. In the design of the monitor the use of very heavy elements is required in order to give a sufficiently low background. Pulses from the monitor were fed to a ratemeter so that the neutron flux could be

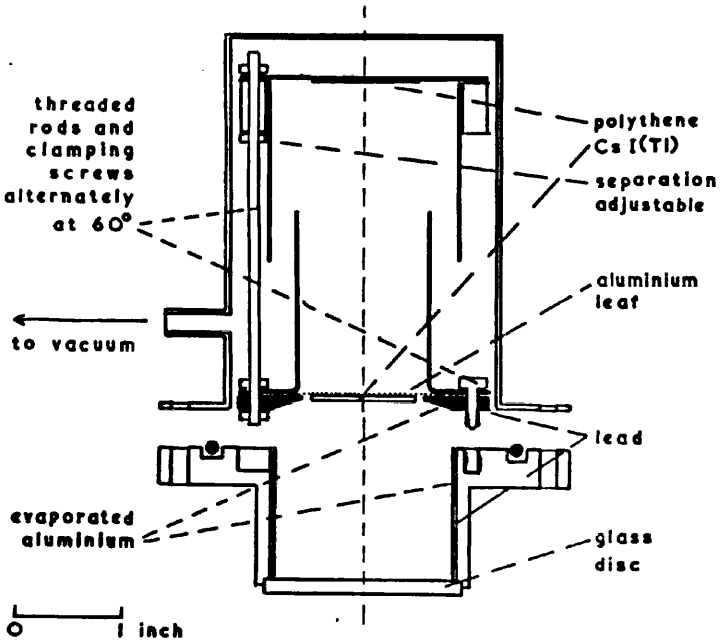


Figure III.1. High efficiency fast neutron monitor of the recoil proton type.

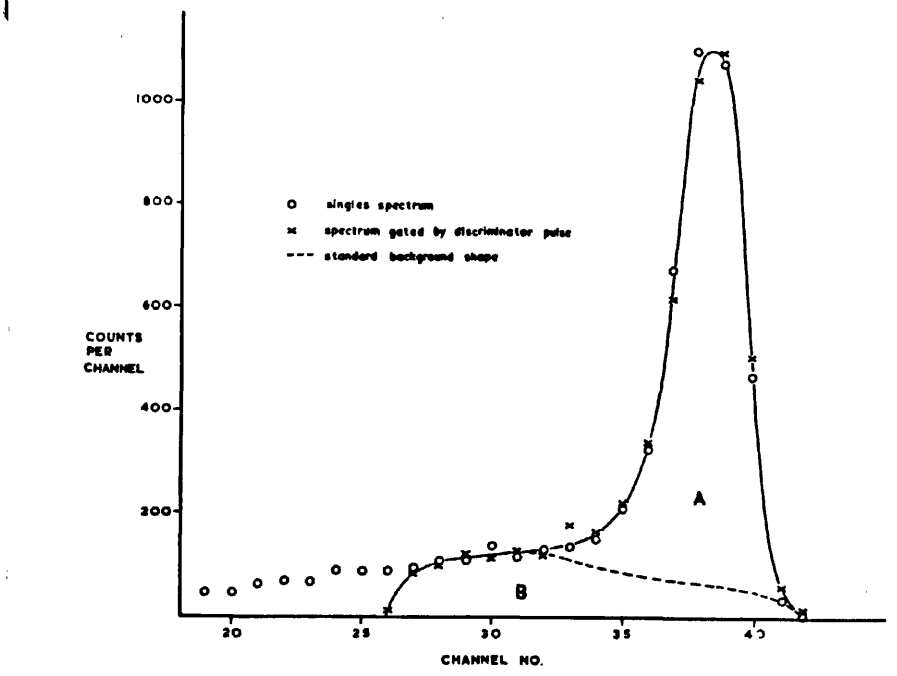


Figure III.2. Pulse height distribution from neutron monitor irradiated with 14 MeV neutrons. O - single spectrum; X-spectrum gated by discriminator; ------ background spectrum, polythene radiator removed.
The efficiency is corrected for the background; the ratio of areas $\frac{A}{A+B} = 0.725$.
The efficiency $\epsilon = 10^{-6}$ when the radiator is 8.5" from the neutron source where ϵ is defined as the number of monitor counts ÷ number of neutrons/unit solid angle.

maintained constant. The neutron energies of 13.5, 14.1 and 15.1 MeV were obtained by making measurements at 0^{o}, 90^{o} and 145^{o} angles to the incident deuteron beam at 220 KeV, respectively. The thickness of the tritium target, which acted as the neutron source, was 70 KV. The corresponding spread in neutron energy was about 0.1 MeV. The end points of the spectra were not used to calibrate the crystal. Instead an independent calibration was made using proton groups from the reactions $^{3}He(d,p)^{4}He$ and $^{10}B(d,p)^{11}B$. Separate tests using a ^{137}Cs source showed that there was no shift in gain when the photomultiplier was changed in position.

III.1(b). Results.

The results of measurements of the energy spectra at the three different neutron energies are shown in figures III.3a,b,c. No correction has been made for edge effects in the crystal but these should be negligible. In these figures the very intense low energy electron background is readily seen. Below 7.5 MeV, this background completely swamps the detection system and it is readily seen that it would inhibit any n-p measurements below this energy. By thinning the CsI(Tl) crystal we can of course reduce this lower energy limit but,as mentioned in Chapter I, using a CsI(Tl) crystal of the minimum thickness required to make

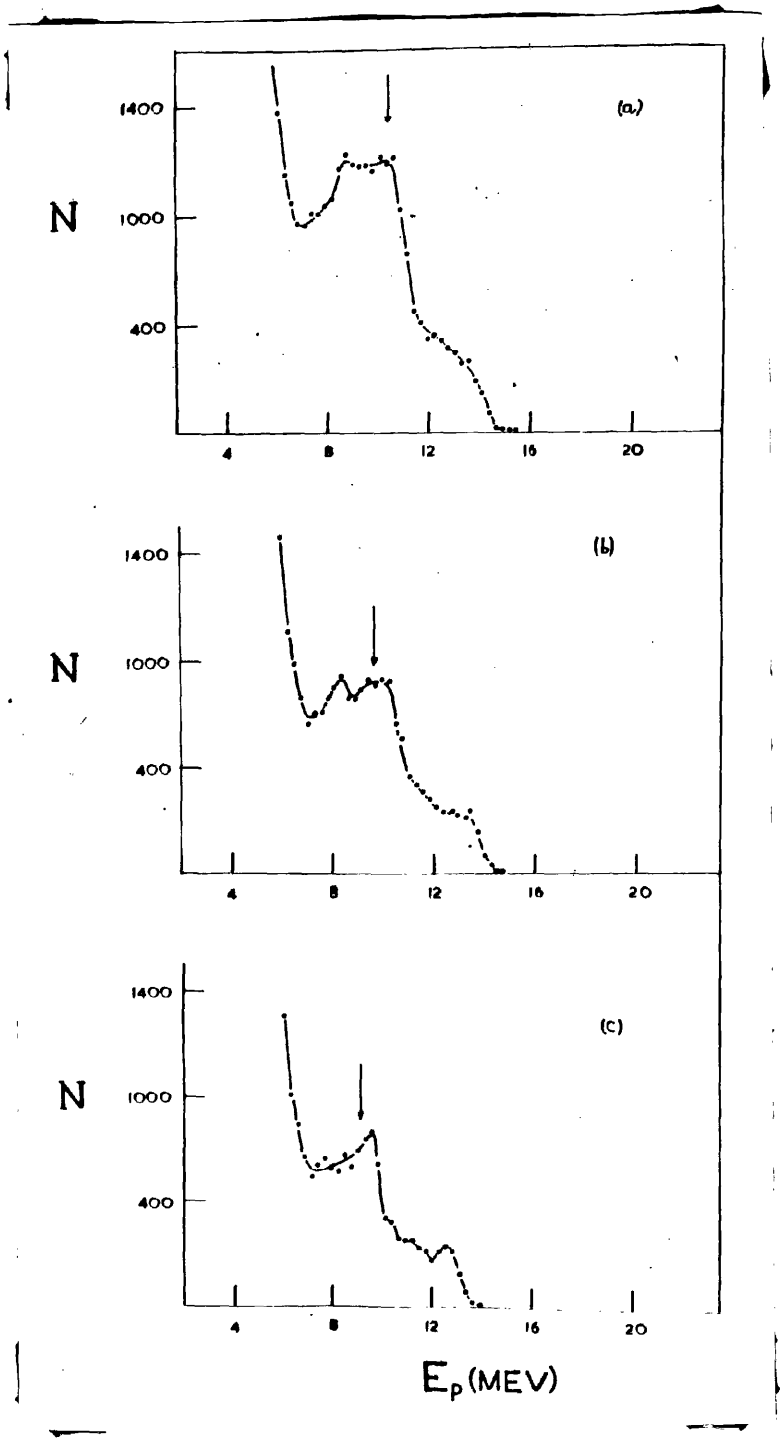


Figure III.3. The energy spectra produced in a CsI(Tl) crystal (1" diam. x 0.4 mm.) on bombardment with fast neutrons. In (a), (b) and (c) the neutrons were of energies 15.1, 14.1 and 13.5 MeV respectively.

n-p measurements over the whole energy range of the emitted particles still results in this intense electron background below 4 MeV.

There is evidence for structure in the spectra. By studying the behaviour of the spectra with incident neutron energy it appears that the structure is most likely to be deuteron groups from the reactions $^{133}Cs(n,d)^{132}Xe$ (Q = -4.14, -4.81 MeV) and $^{127}I(n,d)^{126}Te$ (Q = -4.1, -4.77 MeV). The spins and parities of the levels involved are ^{132}Xe (G.S.0^+, 1st excited state, 2^+), Te^{126}(G.S.0^+, 1st excited state, 2^+), while the spins and parities of the ground states of ^{127}I and ^{33}Cs are $5/2^+$ and $7/2^+$, respectively. The transitions are therefore allowed. Jack (1960) and Colli (1960) find similar structure in the measurement of n-p spectra from medium weight elements and attribute it to n-d reactions. Possible energies of emitted deuterons from transitions to the ground state of ^{132}Xe are indicated by arrows in figure III.3. At an incident energy of 13.5 MeV there is some evidence for a proton group from the reaction $^{133}Cs(n,p)^{133}Xe$.

Values of the total charged particle cross sections for the number of events produced above 7.5 MeV are shown in table III.1. They are in fair agreement with the previous measurements of the partial cross sections in Cs^{133} and I^{127} for n,p, n,α events measured by Coleman et al. (loc.cit.).

Table III.1.

Total Charged Particle Cross-sections, σ, in CsI(Tl) on Bombardment with Neutrons.

Neutron Energy MeV	σ mb.
13.54	16.6
14.11	18.79
15.15	21

In neutron experiments at 14 MeV, it is clear that one of the major difficulties, as discussed in Chapter I, will be to obtain a suitable signal to background ratio. We have seen in Chapter I, that by suitable screening of the CsI(Tl) crystal and the use of very heavy materials in its surroundings, that n-p measurements can be made for energies of emitted protons greater than 4 MeV. Below this energy, however, the very intense electron backgrounds illustrated in figures IIIa,b,c, swamp the spectrum. It is this very intense electron background which the pulse shape analysis technique must eliminate or at least very considerably reduce, to extend the n-p measurements to lower energies. In making measurements on n-α reactions, on the other hand, the thickness of CsI(Tl) required to stop the alphas (5 thou) is very small and as a result we do not have to contend with the low energy electron background. The alpha particles will have energies of a few MeV upwards, according to the Coulomb barrier and the Q values of the reactions, and no electron can produce a pulse of similar magnitude in the very thin crystal. Similarly protons or deuterons emitted simultaneously in n-p events, either from the surroundings or in the CsI(Tl) crystal itself, only produce pulses of energy $\approx$ 6 MeV or less in the crystal. This corresponds to an alpha particle energy of 7.5 MeV. The use of the technique will be

in discriminating against such protons and deuterons and thereby reducing the background in the crystal. The background in the crystal will then consist entirely of alpha particles emitted in n-α events in the crystal itself and the heavy materials used in its surroundings. There will then be a much better chance of obtaining a suitable signal to background ratio in n-α experiments.

We now illustrate the use of the technique in reducing such backgrounds.

III.2. The Reduction Produced in the Low Energy Electron Background Using Pulse Shape Analysis.

In this case we discriminate against electrons. Figure III.4 shows the result of measurements of the low energy electron backgrounds with and without application of pulse shape analysis discrimination. The number of counts in any channel is plotted on a logarithmic scale VS channel number. In this case the bias in the discriminator following the mixer was set so that all electrons with energies greater than 1 MeV produced an anti-coincidence in the gate unit and were accordingly rejected. The CsI(Tl) crystal used was the same as used in obtaining figures III.3a,b,c and was of dimensions 1" by 0.4 mm. This figure shows that with such a bias setting the electron background at 1 MeV has been reduced by a factor of ≈ 40, while at 2 MeV the background has been reduced by a factor

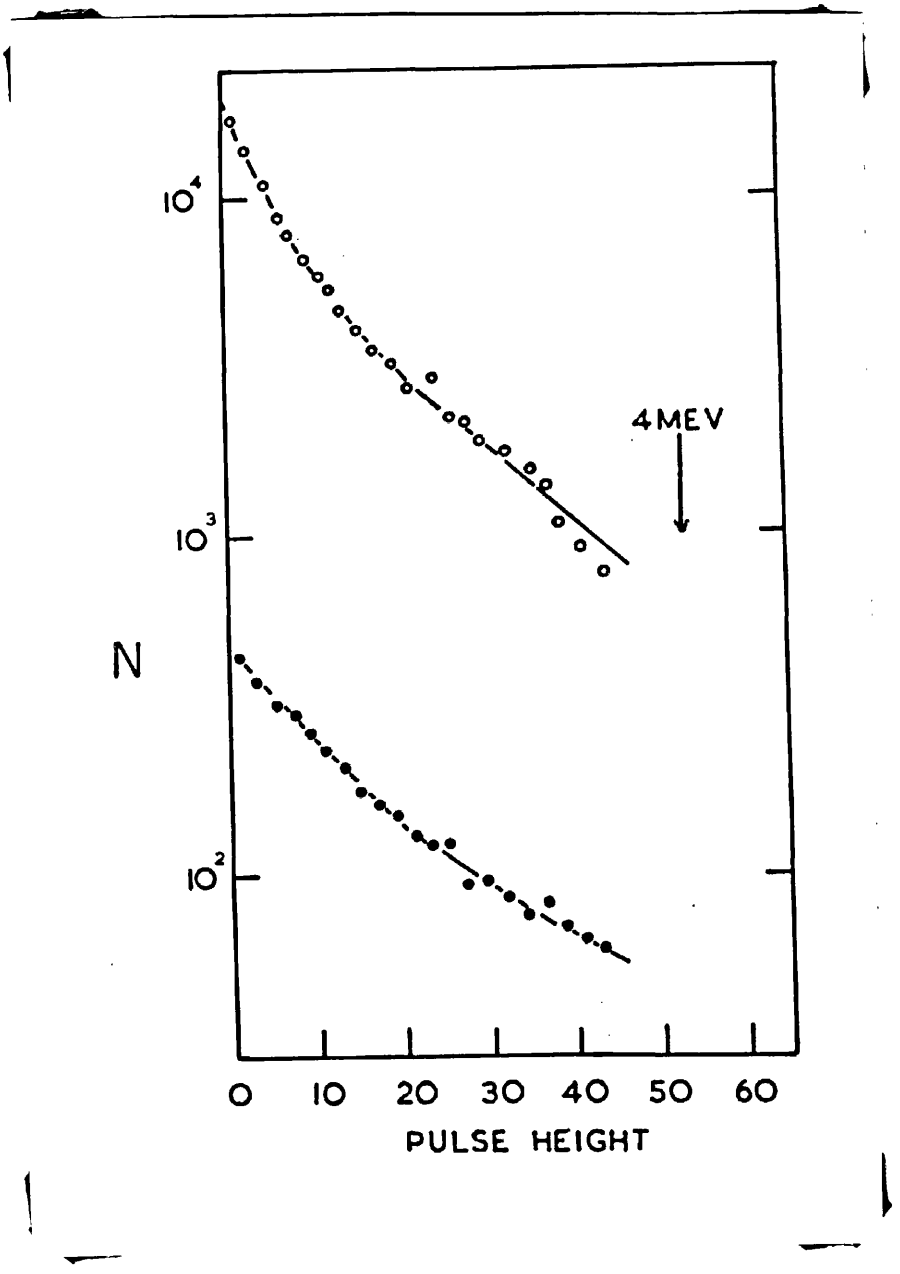


Figure III.4. The low energy electron background between 1-4 MeV produced in a CsI(Tl) crystal (1" diam. x 0.4 mm.) on irradiation with 14.1 MeV neutrons, with and without pulse shape analysis discrimination. The bias setting in the discriminator following the 'mixer' was in this case set to reject all electrons of energy > 1 MeV.

O without pulse shape analysis.

● with pulse shape analysis.

of 20. The total reduction in background between 1-4 MeV is a factor of approximately 30. Figure III.5 shows the results obtained between 2.5-15 MeV using the same bias settings as above. The results obtained when the bias in the discriminator was set to reject all electrons of energies greater than 2 MeV are also presented in this figure. It is seen that above 7.5 MeV, in both cases, the background is unaltered by the application of pulse shape analysis, as expected, since it is then due to protons etc. emitted in the crystal as discussed in section III.1. In both cases the background below 7.5 MeV, the electron background, has been very considerably reduced. In the first case with the bias set to reject all electrons of energies greater than 1 MeV, the background at 2 MeV is reduced by a factor of twenty, in agreement with figure III.4, and at 5 MeV it is reduced by a factor of ten. In the second case the results are in fact twice as good. The reduction at 2 MeV is now a factor of 40 and at 5 MeV a factor of twenty. This is because increasing the bias setting decreases the number of electrons/sec. passing through the circuit and there is a corresponding decrease in pulse pile up. This pile up is a consequence of the general limitation of all such devices that the analysis time involved is necessarily of the order of a few microseconds. Using a ThC^{11} source

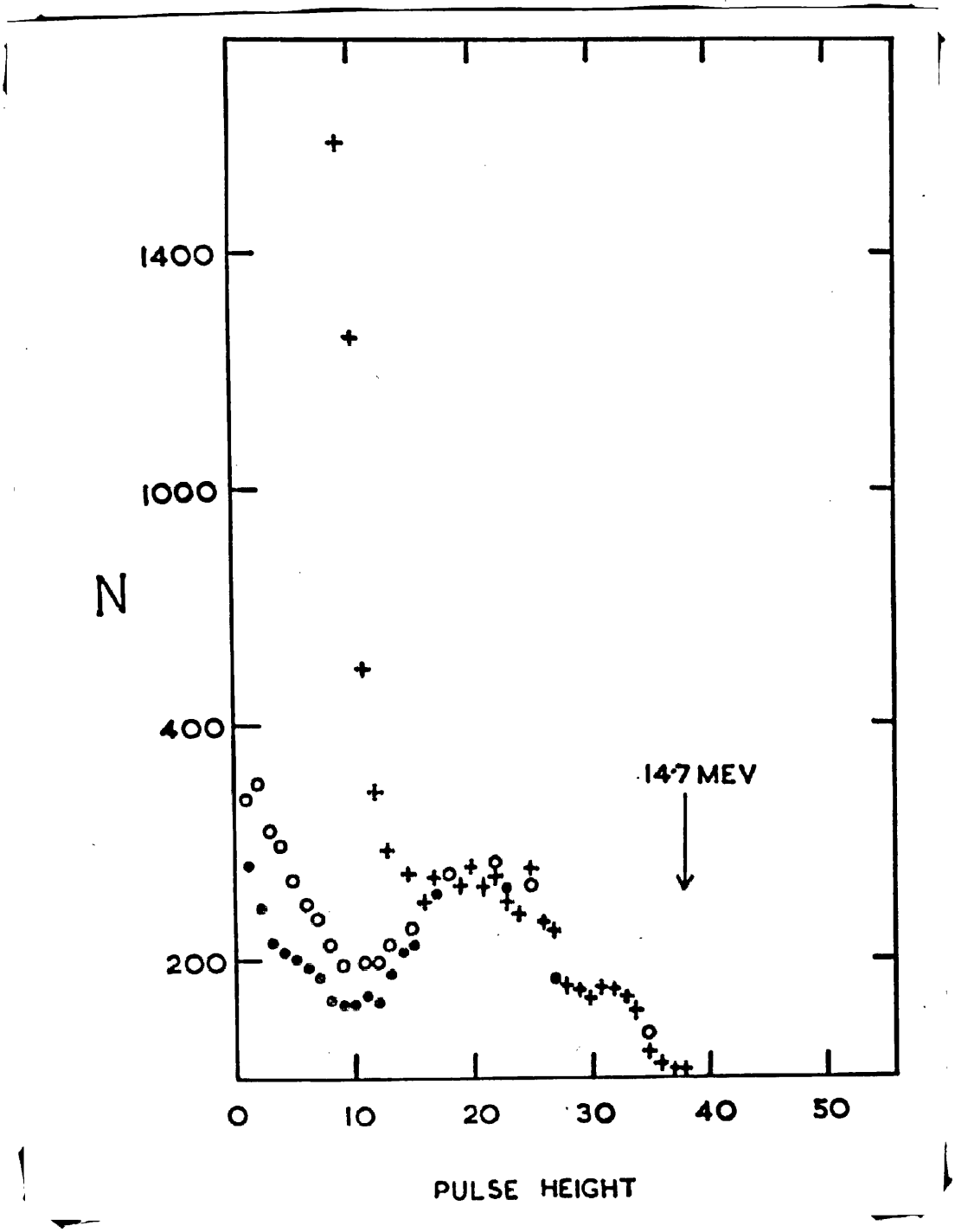


Figure III.5. The energy spectra produced in a CsI(Tl) crystal (1" diam. x 0.4 mm.) on irradiation with 14.1 MeV neutrons with and without pulse shape analysis discrimination.

+ without pulse shape analysis.

o with pulse shape analysis - discriminator set to reject all electrons of energy > 1 MeV.

● with pulse shape analysis - discriminator set to reject all electrons of energy > 2 MeV.

a rejection ratio of 200:1 could be obtained with a counting rate of 100c/sec. In a spectrum of 5923 counts, between 1-2.6 MeV only 30 counts were recorded after application of pulse shape analysis. It is seen from figure III.5 that, with the bias set to reject electrons of energies greater than 2 MeV, the application of pulse shape analysis has reduced the background in the region 2.5-7.5 MeV to the order of half the magnitude of the peak height at 8 MeV of the background due to protons etc., where previously it swamped the crystal.

The reduction in the backgrounds quoted above have been estimated assuming that all counts between 2.5-7.5 MeV were due to electrons. This is possibly not the case and part of the background remaining after pulse shape analysis may be, in fact, due to low energy protons. In this case the results obtained would be better than estimated.

From the results it can be concluded that the incorporation of pulse shape analysis discrimination in a conventional spectrometer proves very successful in reducing the low energy electron backgrounds but that the extent to which such backgrounds can be reduced is dependent on counting rate due to the analysis time involved.

The reduction achieved will, however, be very useful in making measurements on nuclear reactions at low energies of emitted particles.

III.3. The Reduction Produced in the Background due to Protons and Deuterons in the CsI(Tl) Crystal.

In this case we use the technique to discriminate against protons and deuterons. No actual measurements of the decay times of the luminescence produced by deuterons have been made. The similarity of the ionization density produced by protons and deuterons would however be expected to result in the decay times of the luminescence produced by protons and deuterons being very similar. We assume, therefore, that when the technique is used to discriminate against protons that it discriminates against deuterons simultaneously.

Figure III.6 shows the result of measurements of the background with and without application of pulse shape analysis discrimination. The crystal used in this case was very much thinner (200 mg/cm^2) and as a result the low energy background begins at < 3 MeV in energy. The bias setting in the discriminator was set to reject protons of energies equal to or greater than 3.5 MeV corresponding to an alpha particle energy of 5 MeV. It is seen that the application of pulse shape analysis discrimination reduces the background by a factor of eight approximately. Above

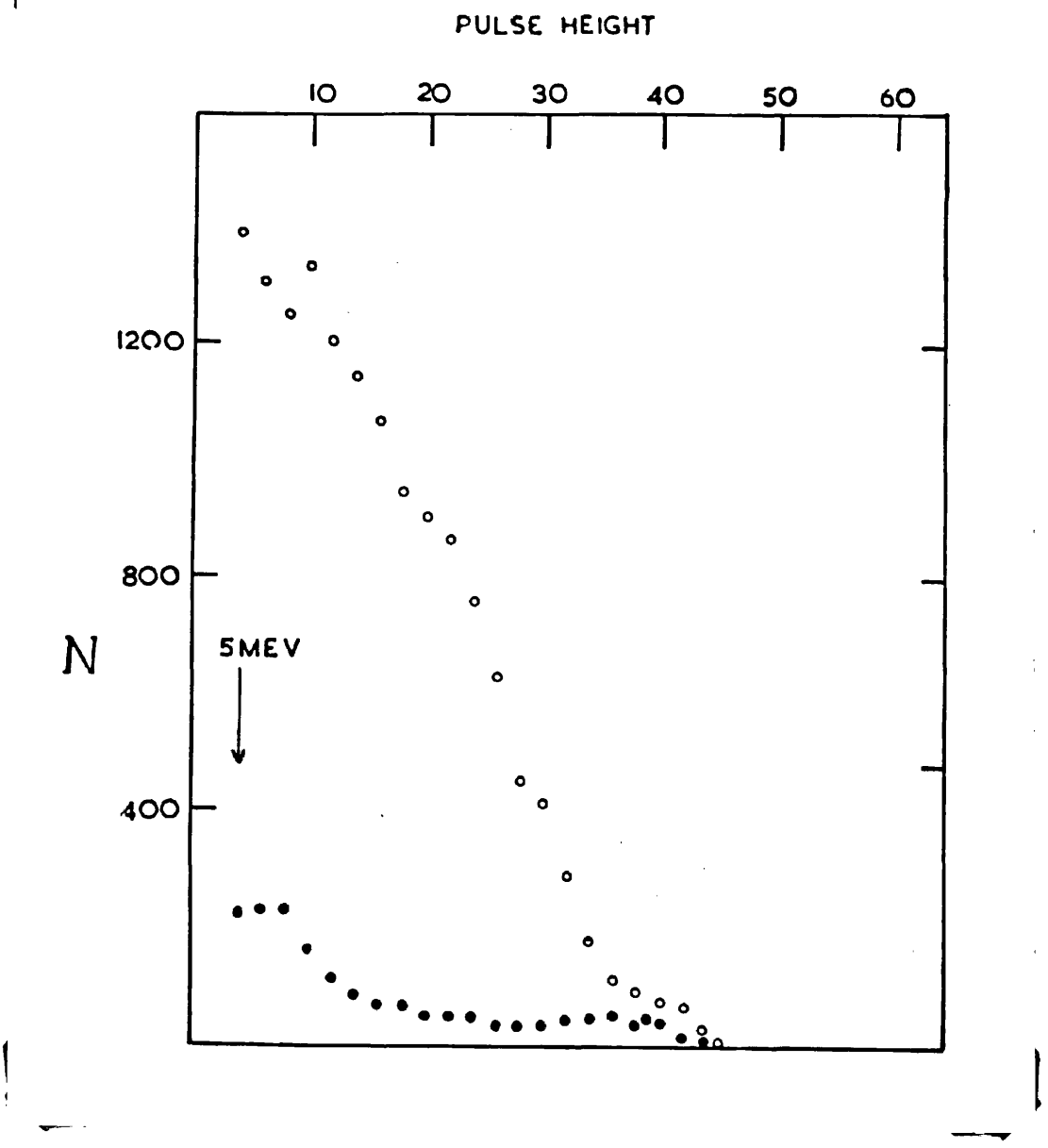


Figure III.6. The energy spectra produced in a CsI(Tl) crystal (1 cm. x 1 cm. x 200 mg/cm^2) on irradiation with fast neutrons.

o without pulse shape analysis discrimination

● with pulse shape analysis discrimination

The arrow indicates the pulse height produced by 5 MeV alpha particles corresponding to a proton energy of 3.5 MeV.

10 MeV, in alpha particle energy, the background, with pulse shape analysis discrimination, is probably the alpha particle spectra from n-α reactions in CsI(Tl). Below this energy we should expect the alpha particle spectra to fall off due to the high Coulomb barrier in Cs^{133} and I^{127} for alpha particles. In fact a rise in the background is observed. This is probably due to n-α events in light elements surrounding the CsI(Tl) crystal. In this case the crystal was mounted in a reflecting cavity and no precautions were taken to avoid such events since we were only interested in illustrating the use of the technique. In a nuclear physics experiment, of course, the crystal would be surrounded with very heavy materials.

Before obtaining figure III.6, tests were made with protons of energy 15 MeV from the reaction $He^3(d,p)He^4$. Absorbers were used to obtain lower energy protons. These tests showed that the rejection efficiency for protons was very high but dependent on counting rate. At 200c/sec. a proton peak at 6 MeV was reduced by a factor of approximately 150. At 40c/sec., in a spectrum of 2500 counts only 5 counts were recorded after application of pulse shape analysis. Tests with alpha particles from Po^{210} (5 MeV) showed that alpha particles were detected with 100% efficiency. In obtaining the results in figure III.6,

the counting rate was approximately 20c/sec.

From the results, we can conclude that the technique is very successful in discriminating against protons and deuterons in the presence of alpha particles. In particular, a considerable reduction in the background in an n-α experiment can be achieved.

III.4. Discussion and Conclusions.

The results presented in this chapter indicate that the application of the technique described in Chapter II produces very considerable reductions in the backgrounds induced in a CsI(Tl) crystal on irradiation with neutrons. This is an important achievment since, as we have seen in Chapter I, one of the major difficulties in neutron physics is to obtain a suitable signal to background ratio. Application of this technique gives a much greater chance of obtaining a suitable signal to background ratio in many experiments. As a result, many experiments which were previously impossible to perform using conventional techniques can now be attempted with every hope of success.

For the reasons given in Chapter I we are primarily interested in using the technique to make measurements on n-α and n-p reactions in intermediate weight elements at 14 MeV. We briefly consider how it will be of use in

these measurements.

(a) n-p reactions in intermediate weight elements.

As we have seen the difficulty in making such measurements using conventional techniques is that below 4 MeV the low energy electron background swamps the detector. An examination of figure III.5 shows that the application of pulse shape discrimination has reduced the magnitude of this background, in the region 2.5-7.5 MeV, to half the level of that in the region 8-12 MeV due to protons and deuterons etc. A consideration of the signal to background ratio obtained by Storey et al. (loc.cit.) in such measurements between 4-15 MeV, indicates that this reduction should be quite sufficient to permit measurements on n-p reactions to be extended to 2.5 MeV. This is especially true when it is remembered that figure III.5 was obtained with a crystal of 4 mm. in thickness and 1" in diameter. We only require a crystal 1 cm by 18 thous. in this experiment so that the reduction produced in the electron background will probably be even greater. The electron background will only begin at about 4 MeV and we will have as a result a much reduced counting rate in the 2-4 MeV region. Again it should be remembered that approximately 40% of all emitted protons have energies between 2-4 MeV, compared with 60% in the region 4-15 MeV over which Storey et al. made measurements. This will

again help the signal to background ratio.

(b) n-α reactions in intermediate weight elements.

In making such measurements the difficulty is to detect the alpha particles in the presence of the proton background. A consideration of figure III.6 shows that this difficulty can be overcome. We are then faced with the problem of detecting alpha particles from external targets in the presence of the alpha particle background in the CsI(Tl) crystal. This is a similar type of problem to that met by Storey et al. in n-p measurements. A consideration of the size of crystal required, the target thickness required and the cross sections etc. show that the experiment has now a very good chance of being successful.

The technique is perfectly general in application and could be used in many branches of low energy nuclear physics. For example, it could be used in making γ-p or γ-α measurements. In the first case to reduce the low energy electron backgrounds, in the second case to discriminate against protons emitted at the same time.

The advantages that the technique has over previous techniques ~~is~~ are self evident since the need for the new technique arose from their limitations, as discussed in Chapter I.

It can be concluded that the incorporation of pulse shape analysis discrimination in using CsI(Tl) as a particle detector represents an important advance in the technique of scintillation counting and is at the same time a useful and valuable instrumental addition.

Future Programme.

It is intended to use the pulse shape analysis discrimination technique to reduce the large electron background at the lower energies in CsI(Tl) and by a similar arrangement to that used by Storey et al. (loc. cit.) to measure the n,p and n,np spectra at the lower proton energies. It is also intended to measure the n-α spectra.

Both the n,np proton spectra and the n-α spectra should be less susceptible to the influence of direct interactions and should be directly comparable with the statistical theory.

The measurements will then be repeated using neutrons of 3-5 MeV from the reaction $^{2}H(^{2}H, n)^{3}H$.

Part II.

Associated Experiments on the Luminescence of Alkali Halide Crystals.

Part II.

Introduction.

Chapter I.

As we have seen in Part I of this thesis, the results of Storey et al. (1958) were important since they indicated a possible method of particle discrimination. Their results were also important, however, in considering the mechanism of the scintillation process. It had not previously been appreciated that the decay time of the luminescence produced in CsI(Tl) by an ionizing particle was dependent on the ionization density produced by the particle, and this fact had not been considered in interpreting the scintillation process. Storey et al. (loc.cit.) presented several possible explanations of their results but before considering these it is convenient first to review the theoretical ideas prevalent at the time of their publication. It should be remarked that only a very small percentage ($\approx 20\%$) of the energy of the incident particle is emitted as luminescence. The bulk of the energy absorbed by the crystal is dissipated in other ways. For example photo-electrons may be emitted, the energy may be used to increase the thermal vibrations of the constituent atoms, or the energy may partly be used in disordering the structure of the crystal.

I.1. Conditions for Luminescence Emission.

It is generally accepted that before luminescence emission can occur a solid must contain atomic or molecular groups, called luminescence centres, in which electronic transitions take place (with resulting luminescence emission) which are protected from interactions with the surrounding material. The potential configuration of such centres have been suggested by Von Hippel (1936) and Seitz (1938). Figure I.1 shows the potential energy of a normal and excited luminescence centre plotted against some configurational co-ordinate x ; in the case of a diatomic molecule x is the separation of the atomic nuclei. All electronic transitions are represented by straight lines. Absorption of a photon raises an electron to the excited state A'. The electron then falls into the position of minimum potential energy in the state B with the thermal dissipation of excess energy. A return to the ground state by the transition BB' gives luminescence. There is then a further loss in thermal energy in going from B' to the state A. Unless the positions of minimal potential energy are in a vertical line, the luminescence quanta will always be of smaller energy than the absorbed quanta, so the absolute energy conversion efficiency will in general be less than unity. If the curves for the two states come sufficiently close together, as shown, then at high

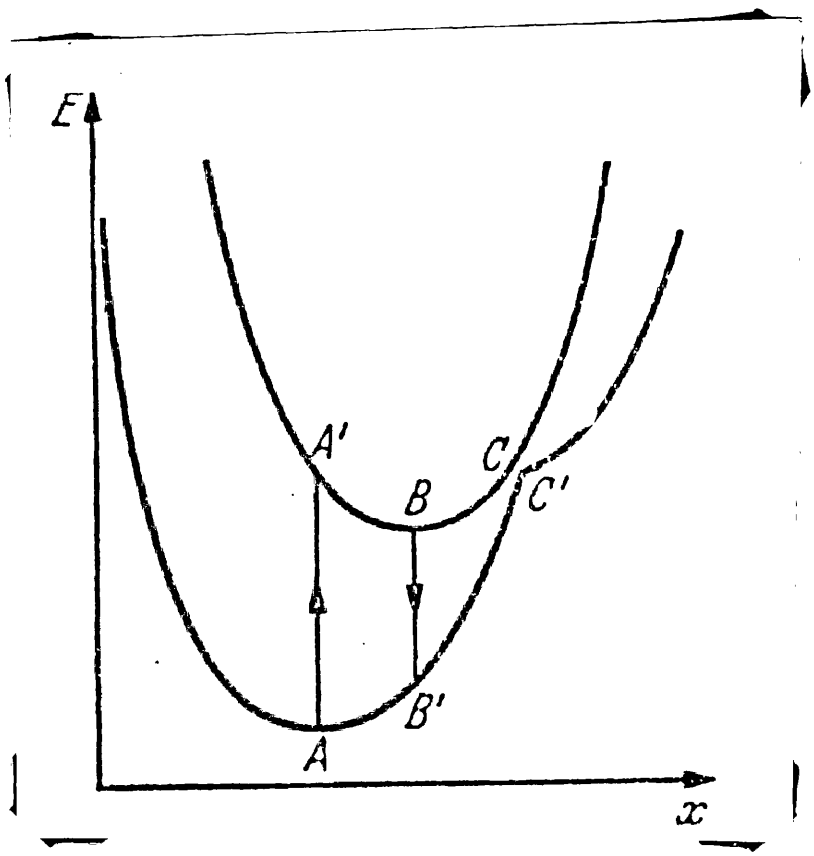


Figure I.1. Potential energy configuration of a normal and excited luminescence centre.

temperatures the excited centre returns to its ground state by a series of vibrational interactions with its surroundings, by the path CC'B'A. No luminescence then occurs. During these processes, it is assumed that the configuration co-ordinates of the centre remain fixed. The efficiency for luminescence clearly depends on the relative probabilities of electronic or vibrational transitions in the centre.

I.2. Energy Transfer in Crystal Phosphors.

In using phosphors as scintillators we are never concerned with absorption and emission processes entirely confined to the luminescence centres. Charged particles lose their energy to the lattice producing considerable ionization. It is necessary to know how this energy is transferred to the specific centres where luminescence occurs. To do this the modern picture of the energy states of electrons in the crystal lattice is used. In ionic crystals the forces which exist between ions cause a convergence of the individual molecular ~~events~~ ORBITS of the valence electrons into diffuse bands belonging to the entire crystal.

The final processes involved in luminescence are essentially confined to the valence electrons and the energy band system for these electrons is given in figure I.2. In such crystals the highest occupied band is usually completely filled according to the Pauli Exclusion principle.

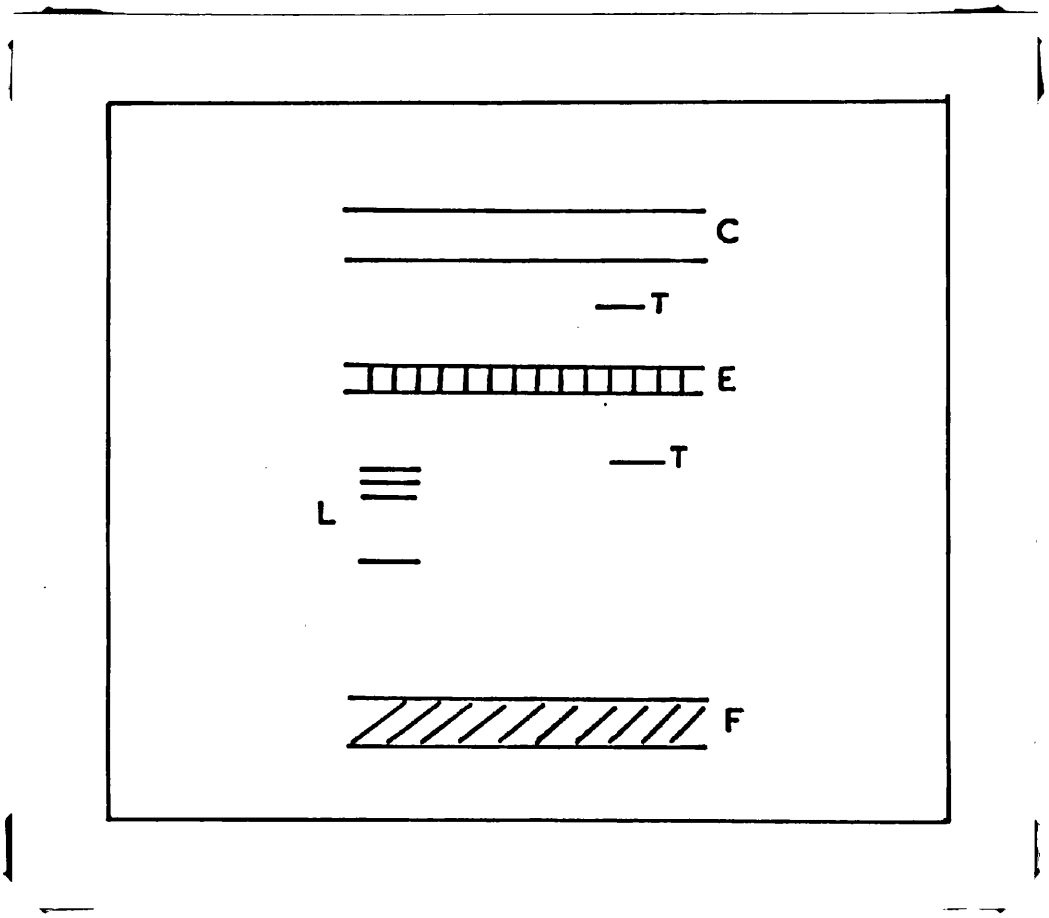


Figure I.2. The energy band system in ionic crystals, for the valence electrons.

Defects and impurity atoms or excess lattice constituents give rise to rather more discrete levels which are localised at the defect or impurity atom in the lattice. These can trap electrons which have been raised to the empty band above. The energy states of the luminescence centres lie in the normally forbidden region between the filled and empty energy bands. The process of excitation of the crystal by ionizing particles can be described thus in terms of such a model.

After excitation we are left with the situation where numbers of electrons of relatively small energies remain in the empty band of figure I.2. The associated vacancies in the valence band behave like positive charged electrons and so are known as positive holes. These holes capture electrons from the ground states of the luminescence centres and also from filled trapping levels. Then electrons in the empty conduction band can combine with empty luminescence centres or be captured in trapping states. The electrons in the conduction band can thus return to their normal ground state either via traps or luminescence centres. The combination with luminescentecentres, however, results in tho emission of the electron energy as light in its transition to the ground state, whereas transitions of trapped electrons to combine with positive holes in the valence band will be non-radiative. Absorbed quanta may also excite

the crystal without ionization (band E). Again excitation energy is not localised but moves rapidly through the crystal in the form of excitation waves. These excitation waves also exhibit corpuscular properties; the associated particle is referred to as an exciton. The exciton consists of a positive hole and electron revolving around each other in closed orbits. The exciton may arrive at a crystal imperfection and radiate its energy as elastic waves or it may be captured by an impurity atom, causing the impurity to become excited. It is through this process that excitation waves may be important to the scintillation process.

It is clear that the luminescence efficiency depends on the relative number of luminescence centres and non-radiative centres and on their respective cross sections for capture of electrons. Again it depends on how much of the particle energy used in creating excitons is converted into luminescence. Finally, as we have seen, it depends on the probability for radiative and non-radiative processes in the luminescence centres.

The rise and decay times of the luminescence in inorganic phosphors clearly depends, on this picture, on the time delay between absorption of the exciting radiation and the transfer of energy to the luminescence centres, and also on the probability of radiative transitions within the

centres. If the transport time is small then the decay will depend solely on the probability of radiative transitions within the luminescence centres. This transport time is clearly dependent on the probability for thermal release of the electrons from the electron traps. If this probability is very low then phosphorescence occurs. The dependence of phosphorescence on trap depth and distribution has been developed by Randall and Wilkins (1945). In this thesis we are concerned only with prompt emissions $< 10^{-4}$ secs.

I.3. More Specific Explanations of the Luminescence Processes.

In the above section we have reviewed the general ideas presented by Garlick (1952) and Swank (1954) on which most explanations of the scintillation process in inorganic crystals are based. In this section we consider the more specific models presented in explanation of observed experimental results in the scintillation process.

Rossel and his co-workers in a series of papers:- Bonanomi and Rossel, 1952; Hahn and Rossel, 1953(a) and (b); Enz and Rossel, 1958; and Chauvy and Rossel, 1960; have shown that several components in the decay of the luminescence produced in several alkali halide crystals, by ionizing particles, can be explained on the following model. The luminescence centres occur in the undisturbed lattice. The potential configuration is shown in figure I.3. De-excitation

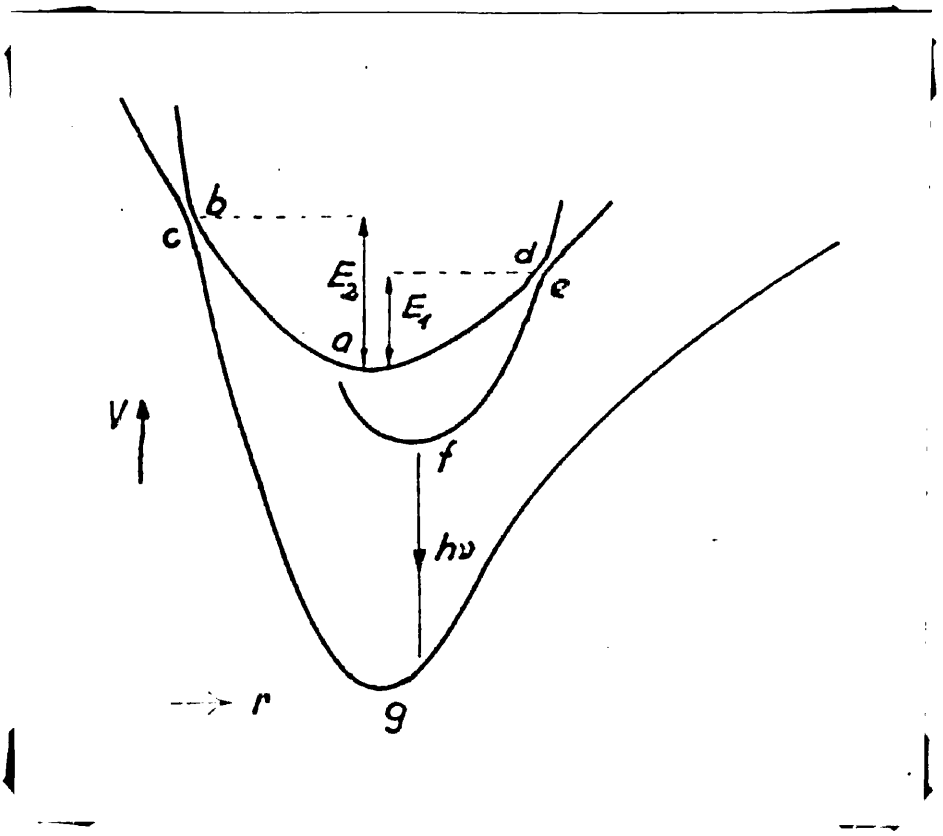


Figure I.3. The potential configuration, suggested by Bonanomi et al., for the luminescence centres in alkali halide crystals.

of the centre occurs in two ways; radiatively by adefg and non-radiatively by abcg with activation energies E_1 and E_2. Then $\frac{1}{\tau} = K_1 + K_2$ where K_1 and K_2 are the probabilities for the two processes. $K_1 = S_1 \exp(\frac{E_1}{kT})$ and $K_2 = S_2 \exp(\frac{E_2}{kT})$, where S_1 and S_2 are constants, k is Boltzmann's constant, and T is the absolute temperature of the crystal. τ is the value of the decay time component and should become larger as the temperature of the crystal is lowered. E_2 is much greater than E_1 and consequently non-radiative transitions only become important at high temperatures. The luminescence efficiency $\eta = \frac{K_1}{K_1 + K_2}$ and should increase as the temperature of the crystal is lowered. The components of the decay of the luminescence which obey this model are referred to as the 'cold' components. From plots of the logarithm of τ versus $\frac{1}{T}$, it is possible to obtain values for E_1 and E_2. Other components are found, however, which do not obey this model. These are referred to as the 'hot' components. It is concluded that the luminescence centres for these components are due to crystalline defects produced in the crystal either by the addition of thallium or by thermal treatment. In this case the luminescence mechanism can only be explained by a complicated mechanism. In this work no variation in the decay time of the luminescence was observed with ionization density.

Eby and Jentschke (1954) find that in NaI(Tl) the main characteristics of the luminescence are a rise time of 5.9×10^{-8} secs. and decay time of 0.23 μ.sec. The rise time is independent of particle type and thallium concentration but the decay time is dependent on the thallium concentration, although again independent of particle type. Upon excitation by a particle, it is assumed that a number of excited states are created in the crystal. The decay time of these states can be observed by studying the decay of the luminescence produced in the crystal. To explain their observations Eby and Jentschke suggest that a minimum of two excited states is required:- a radiative state in the luminescent centre and a non-radiative state feeding it(not necessarily associated with the luminescent centres.) The transport time of the energy lost by the exciting particle to the states is assumed to be negligible and to have no effect on the observed luminescent decay times. It is not clear which time is associated with the radiative state but from the behaviour of the scintillation efficiency with thallium concentration it appears most likely to be the rise time. In crystals containing low thallium concentrations (<0.00018 mole fractions of thallium) an additional fast spike (1.2×10^{-8} secs.) is present in the decay, interpreted as being due to the decay of an exciton. In crystals of high thallium concentration (>0.00095 mole

fractions of thallium) an additional fast component is present in the decay (1.5×10^{-8} secs.). This is interpreted as being due to another excited state of the luminescence centres. Apparently the difference of the scintillation efficiency for different particles, i.e. with different ionization density, must be explained on the basis of the probability for exciting the radiative and non-radiative states during the initial energy transfer in the crystal. It should again be noted that no variation in the decay time characteristics of the luminescence was observed with ionization density.

Mateosian et al. (1956) find that the scintillation efficiencies of alpha particles in pure and thallium activated crystals of NaI become approximately equal at low temperatures. At room temperatures, of course, the scintillation efficiency in the thallium activated crystal is much greater. This effect has been observed by Van Sciver (1956), and Knoepfel et al. (1957) have observed a similar effect in CsI crystals. It is suggested that this effect is due to the presence of two different luminescence centres in the crystals; one present in the pure crystal, the other being induced by the thallium. The luminescence centre in the pure crystal is similar in behaviour to that described in section I.1. As the temperature of the crystal is lowered the internal

luminescence efficiency increases due to the reduced probability for non-radiative transitions. The second type of centre is, following Seitz (1939), formed by two adjacent thallium ions. In this centre the activation energy for non-radiative transitions is less than that for radiative transitions. Thus, unlike the first centre, the efficiency decreases as the temperature is lowered. Both types of centre are present in the thallium crystal so that the luminescence efficiency at any one temperature is a combination of these effects. At low temperatures it is assumed that the transfer of energy from the crystal lattice to the thallium centres is inhibited so that the scintillation efficiencies in the pure and thallium activated crystals become equal.

Since the early measurements on inorganic phosphors it had been known that the luminescence efficiency for alpha particles was less than that for electrons. Allison and Casson (1953) suggested that this was probably due to the increased importance, in the degradation of ion energy, of the direct transfer of momentum to the atoms of the crystal. Mateosian et al. (loc.cit.) find however that at $-150^{\circ}C$ in a pure NaI crystal the luminescence efficiency for electrons becomes greater than that for alpha particles. Allison and Casson's argument cannot account for this, and in fact only Eby and Jentschke's postulate of different

probabilities of exciting different luminescent states by different particles can yield an explanation.

Recently Meyer and Murray (1960) have calculated the response of thallium activated alkali halide crystals to charged particles on the basis of a model which treats the scintillation process in three parts: (1) slowing down of the incident charged particle with its energy going into the formation of electrons and holes, either free or bound together as an exciton; (2) the transport of at least part of this energy to activator sites, raising the activator to an excited state; (3) radiative decay of the excited state. The light output is assumed to be directly proportional to the number of energy carriers arriving at activator centres, no further consideration is given to the decay process itself. They deal with only parts (1) and (2). Following Van Sciver (1956) they assume that an electron and a positive hole must be captured by the activator, either in succession or as an associated pair (exciton). Thus the electrons and holes produced by the incident primary particle must recombine to stimulate an activator site. These authors consider in detail the case of recombination in the wake of the particle followed by diffusion of excitons. Good agreement with experiment is obtained. The model predicts the scintillation efficiency

as a function of ionization density, concentration dependent effects and a diffusion length for the carriers. A similar type of picture had been presented by Van Sciver (1956) in explaining his results on the behaviour of the emission spectra of NaI with ionization density, temperature and thallium concentration.

I.4. <u>In CsI(Tl) the decay time τ of the luminescence depends on the ionization density of the exciting particle.</u>

In the above interpretations of the scintillation process, no consideration was given to the fact that the decay time of the luminescence might be dependent on the ionization density produced by the particle. The results of Storey et al. (loc.cit.) showed this to be the case in CsI(Tl) and we now consider the possible explanations of this effect, which they suggested.

Comparing the results of Knoepfel et al. (loc.cit.) for the variation of the decay time τ with temperature and their results for the variation of τ with ionization density, Storey et al. suggest that the ionization density produced by a particle determines the effective local temperature at which emission occurs. Their results can then be explained in terms of a simple model employing two sets of electron traps with different energy levels. Decay of the trap occurs when it receives enough thermal energy to reach an excited state, decay from which leads to luminescence.

It is supposed that the higher energy trap is responsible for the short lifetimes found in the decay, while the lower energy trap gives rise to the common long component found in the decay. The difference in the two lifetimes is due to the energy difference between the trap and the levels responsible for the luminescence. It would be expected that the trap with the smaller energy difference would be more susceptible to temperature changes than the other one. This would lead to shorter lifetimes for particles producing greater ionization density if different ionization densities correspond to different local temperatures. A comparison of the light emitted in the long component to the total light output suggests that the lower trap, responsible for the long component, is fed from the higher one in competition to the radiating state.

This interpretation would lead one to expect a greater difference in the decay times of the luminescence for different particles as the temperature of the crystal is lowered.

Another quite different interpretation of the results is that a variation in ionization density results in a different distribution in the initial population of radiating states. The observed variations in the value of γ would

then reflect the different contributions of many radiating states with different decay times. The simplest and most extreme case would be two radiating states corresponding to the values of decay time observed for alpha particles and electrons. The order of magnitude of the values of decay time observed does however seem to be more consistent with those expected for traps rather than for radiating states.

The results indicate that in CsI(Tl) the efficiency and the decay times of the luminescence vary with the density of ionization. It might be thought that both effects might arise from the same cause. Storey et al. considered the possibility that quenching mechanisms occurring during the transport times to the emitting centres are responsible. In this case the observed variation of τ with ionization density is attributed to a transport (or recombination) mechanism (Curran, 1953), and slow quenching occurring during the transport time to the emission centres results in a greater decrease in efficiency for smaller ionization densities, which correspond to longer decay times. Since, however, the observed efficiency of CsI(Tl), and LiI(Eu), Schenk and Neiler (1954), appears to first increase and then decrease with ionization density, it is necessary in addition to have a fast quenching mechanism of the type observed by Kallman and Brucker (1957) in a wide

variety of phosphors. This fast quenching is completed before any appreciable quantity of light is emitted, and leads to a monatonic decrease in efficiency with ionization density.

I.5. The function of the activator in the scintillation process.

Clearly, the part played by the activator in the crystals must be understood before any explanation of the scintillation process is complete. Does the activator act as the luminescence centre or is it associated with the non-radiative processes in the crystal?

Bonanomi and Rossel (loc.cit.) examining the components found in the decay of the luminescence, the emission spectrum, and the efficiency of the luminescence in crystals with or without thallium as activator, conclude that the thallium is not associated with the emission process but with the non-radiative (or trapping) processes in the crystal.

Studying the effect of thermal treatment of pure CsI crystals, the effect of heating a pure crystal in sodium vapour, and the effect of adding thallium to the crystal, by examining the emission spectra and the behaviour of the decay time of the luminescence with temperature, Knoepfel et al. (loc.cit.) conclude that the function of the thallium is to produce iodine vacancies in the crystal lattice which then act as the luminescence centres.

Eby and Jentschke (loc.cit.) examining the main

components present in NaI(Tl), find that, irrespective of whether the rise of the luminescence or the decay of the luminescence is associated with the radiative state, the initial population of the radiating state increases as the thallium concentration in the crystal increases. They conclude that the thallium must be associated with the luminescence centres, or be the luminescence centres.

It could be that the thallium radiates energy transferred non-radiatively to it from some other centre, in a sensitised luminescence process, as suggested by Dexter (1953).

The response of thallium activated alkali halide phosphors to excitation by light was interpreted by Seitz (loc.cit.) in terms of the thallous ions which were assumed to occur substantially in the lattice and which acted as the luminescence centres. Further papers by Williams (1951, 1953) and Knox and Dexter (1956) among others have been given interpreting the emission and absorption spectra of K Cl(Tl). Ewles and Joshi (1960) have pointed out that there are several contradictions in this work and that further research in the field is necessary. They conclude that in K Cl(Tl) in fact there are three types of luminescence centre associated with the thallium.

In considering the part played by the activator in the luminescence process we must consider the concentration

quenching mechanisms suggested by Johnston and Williams (1950), and Kallman and Furst (1950). These result in a decrease in the luminescence officiency due to the presence of too much thallium. Meyer and Murray (1960) suggest that a reduction in the luminescence efficiency with ionization density is due to insufficient activator being present. This is apparently in contradiction with the concentration quenching mechanisms.

I.6. Present Programme.

We have seen, in the preceding sections, that only Storey et al. (loc.cit.) had given any consideration to the possibility of the decay time of the luminescence being dependent on ionization density. No previous measurements on the decay times of the luminescence in inorganic phosphors had suggested that the ionization density produced by the exciting particle had any influence on the decay time, although such a dependence had been noted for the luminescence officiency. It was possible that it was only in CsI(Tl), that the characteristics of the energy transfer were such that the decay time of the luminescence is dependent on ionization density. Before any definite conclusions on the mechanism of the scintillation process could be reached it was clearly necessary to make more extensive measurements on other thallium activated alkali

halide crystals to see if a similar effect is observed in these. Accordingly, a systematic study was made in the commonly available alkali halide crystals, of the dependence on ionization density, of the decay time τ of the luminescence, and the efficiency of the luminescence. The results are presented in Chapter II.

Storey et al. (loc.cit.) suggested that the variation in the decay time of the luminescence with ionization density might be due to a local heating of the crystal. It is obvious that more information could be obtained about this effect by measuring the decay time of the luminescence at different temperatures. In Chapter III we present measurements on the dependence on temperature of the decay times and efficiencies of the luminescence produced in CsBr(Tl) and CsI(Tl) by alpha particles and protons.

The part played by the activator in the luminescence processes is not fully understood, as we have seen in section I.5. Thus measurements were made in CsI(Tl) on the dependence on thallium concentration of the decay times and efficiencies of the luminescence produced by particles of varying ionization density. The results of these measurements are given in Chapter III.

All of these measurements were of importance from the point of view of the particle discrimination technique discussed in Part I of this thesis. It was possible that a more suitable crystal than CsI(Tl) could be found for use in this technique, in which the decay time of the luminescence was more dependent on ionization density. Variation of the temperature and thallium concentration of the crystals might produce a greater difference in the decay times of the luminescence produced by different particles.

Part II.

Chapter II.

The Luminescent Decay of Various Crystals for Particles of Different Ionization Density.

In the scintillation counter the light produced by an ionizing particle in a luminescent material is converted into an electrical current pulse with the same time variation. As mentioned in Chapter I, measurements by Storey, Jack and Ward (1958) indicated that in CsI(Tl) the decay time of the luminescence was dependent on the average ionization density ϱ due to the ionizing particle. In Part I of this thesis we have described a technique which employs this difference to distinguish between alpha particles and electrons, electrons and protons, and protons and alpha particles. It is of practical as well as theoretical interest to see whether this difference in decay time is found in any other crystal and, if so, whether any other crystal would be more suitable for practical use.

II.1. Experimental Method.

Measurements of the decay times of the luminescence produced by alpha particles, protons of varying energies and electrons have been made in LiI(Eu), NaI(Tl), KI(Tl) and CsBr(Tl). Measurements have also been made on a standard Harshaw CsI(Tl) crystal and compared with the

results of Storey et al. (loc.cit.), to test the method used. All the crystals were obtained from the Harshaw Chemical Company and had a molar concentration of 0.1% of thallium, except the LiI(Eu) crystal. The molar concentration of europium was in this case 0.1%.

The crystals were mounted directly on to the face of a Du Mont 6292 photomultiplier with silicone grease. A highly reflecting aluminium cylinder with an end window of thickness 1 mg.cm^{-2} was used as a light reflector. The crystal was then excited by the entry of the particular particle in which we were interested.

The 'Voltage pulses', or integrated current pulses (the leakage time constant being 865 μ.sec.), were fed through a cathode follower and matched delay to a Tektronix Type 541 cathode ray oscilloscope. Measurements were made on a short and long time scale in each case, the length of the delay line being chosen according to the length of the decay time to be measured (this had been estimated from a provisional measurement). Photographs were then taken of the individual pulses produced by each type of particle.

The advantage of using the 'voltage pulse' is that charge fluctuation is not so troublesome as it is in the case of the current pulse. The 'voltage' pulse averages the statistical variations on the 'current' pulses.

The time base speeds on the Tektronix oscilloscope were calibrated using a crystal controlled standard frequency oscillator, Furzehill Type 9415. The probable error in the calibration was ± 1%; the measurements were corrected according to this calibration.

^{137}Cs gamma rays (E = 661 Kev) were used to produce photo-electrons in the crystal; a ^{210}Po source (5.3 MeV) was used to give the alpha particles. High energy protons were obtained from the reaction $^{3}_{2}He(d,p)^{4}_{2}He$. Lower energy protons were obtained using aluminium absorbers. Very low energy protons were obtained from the $^{2}_{1}H(d,p)^{3}_{1}H$ reaction. In order to keep the pulse height on the oscilloscope constant, various values of the photomultiplier voltage were used. The change in photomultiplier gain at these different voltages was later measured using a Hutchinson-Scarrott 100-channel pulse height analyser. Knowing the difference in gain at the voltages used, all the graphs of the voltage pulses could be normalised to the same gain.

An alternative method of studying the luminescence is to observe the emission spectra of the light with a suitable spectrometer. The difficulty in this method is the very small amount of light emitted but it has the advantage that all the emitted light can be observed and the method is not limited, as is the method described above, by the spectral response of the particular photomultiplier

used. This is not a serious disadvantage since in the luminescence of thallium activated alkali halides most of the emitted light is in the spectral region (3000-7000°) matching the response of the commonly available glass window photomultipliers. (Van Sciver, 1956; Hrehuss, 1960) and Knoepfel et al., (1957). We chose to use photomultipliers since the experiments on the luminescence were performed partly with a view to improving the technique described in Part I.

II.2. Analysis.

II.2(a) Decay Times.

Two methods of analysis were used.

In the first method, similar to that used by Storey et al. (loc.cit.), the photographs of the 'voltage' pulse were projected onto a large screen and the recorded pulses carefully traced on to fine graph paper. The graphs were corrected for the leakage time constant of 86.5 μ.sec., and then carefully differentiated to obtain the shape of the original current pulses. This provides a consistent method of averaging the statistical fluctuations in the current pulse. Plots were then made of these current pulses on semi-log. paper, plotting the pulse height I at time t vs t. If these plots are straight lines it means that the decay of the luminescence can be represented

by a single exponential component. In fact it was found that only for LiI(Eu) was this the case. In all other cases two components were found in the decay. This is illustrated in figures II.1,2. Figure II.1 shows the plot of log I vs.t for LiI(Eu), where $I = \frac{dv}{dt}$, obtained from a voltage pulse produced by a 15 MeV proton. The single exponential nature of the decay is illustrated. Figure II.2 shows a similar plot for CsBr(Tl). The long component present in the decay is readily seen. By subtracting off the amount of long component present in the decay, the value of the short component is readily obtained.

In practice, several photographs were taken of the voltage pulses with various values of time scale on the oscilloscope. The photographs on the longest time scale provided the plots from which the long component of the decay of the luminescence was measured. The ratio of the amount of light in it to the total light output was also obtained from these plots. The value of the short component present in the decay was determined from similar plots obtained from photographs on a much faster time scale. The long component was superimposed on these plots and carefully subtracted off. This left the short component, the value of which was then measured.

The accuracy of the measurements of the short component clearly depends on the accuracy with which the value of

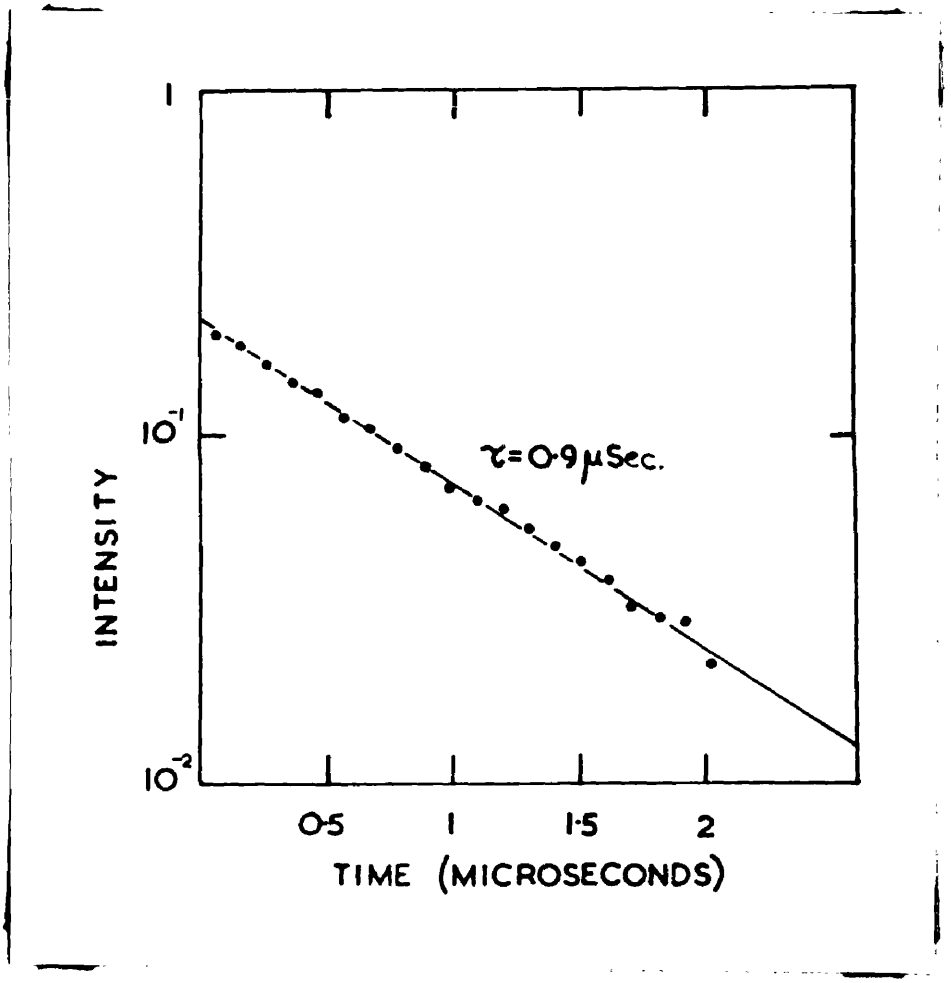


Figure II.1. The Decay Curve of the Luminescence produced by 15 MeV protons in LiI(Eu).

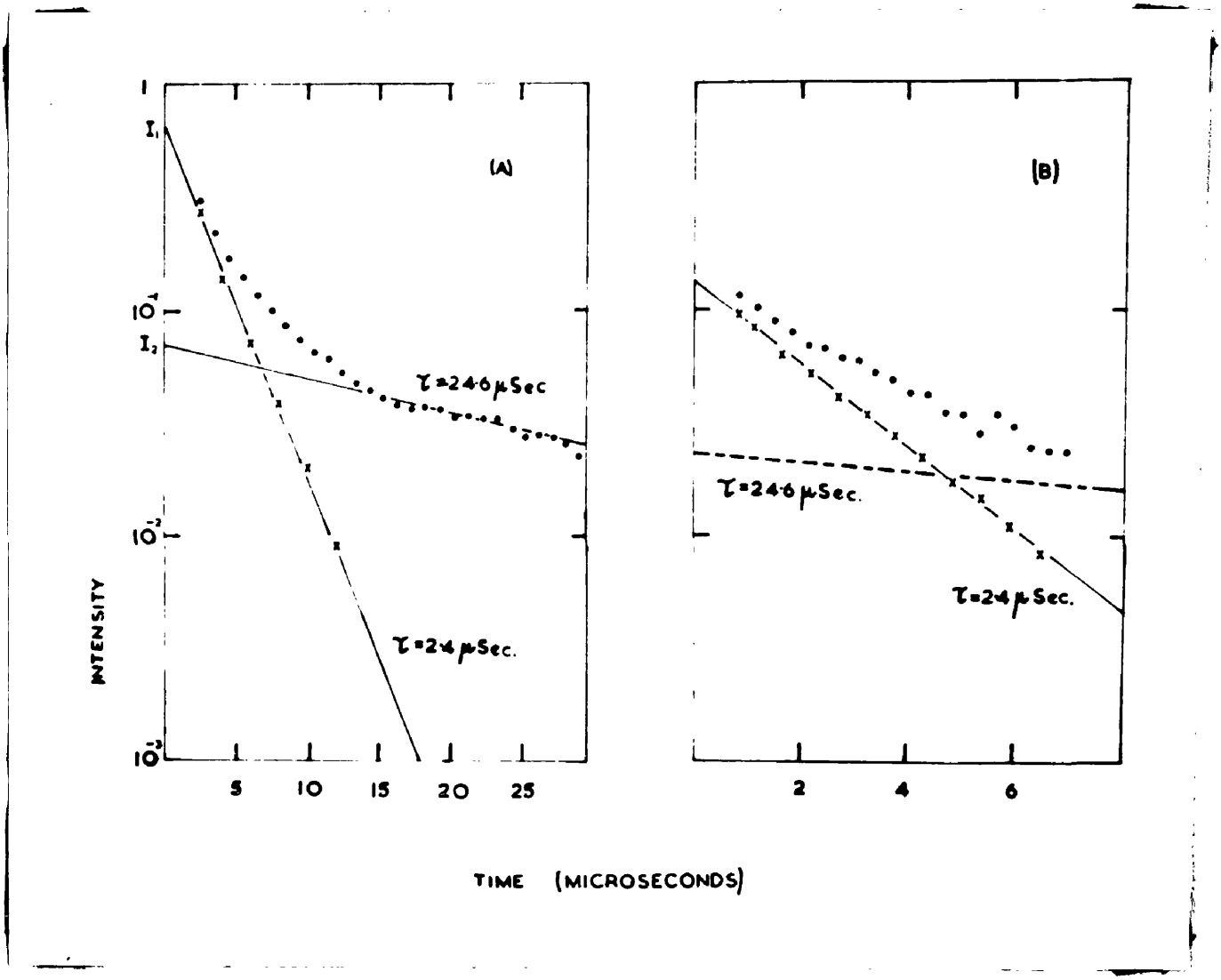


Figure II.2. The Decay Curves of the Luminescence produced by 15 MeV protons in CsBr(Tl).

the long component is known, and how much of this component is present in the decay. For this reason, at least three measurements of the value of the long component and the amount of it present in the decay were made. The greater in value is the long component than the less is its effect on the value of the short component. Similarly the smaller its intensity in the decay, the less is its effect on the value of the short component.

It should perhaps be remarked that this method of analysis is not only applied to inorganic phosphors but has been applied by Owen (1959), among others, to organic phosphors. Here again short and long components are found in the decay of the luminescence.

From the practical point of view the shape of the pulse over the first few microseconds is of importance rather than the actual components present in the decay. For this reason, in the second method of analysis, the values of the decay time τ', over four lifetimes of the short component as measured by the first method of analysis, were measured, assuming the decay to consist of only one exponential component. The time involved in this method of analysis is of course much shorter.

II.2(b). $\frac{Imax}{E}$, R.

Values of $\frac{Imax}{E}$, the relative peak current/unit energy, and of R, the ratio of the light output in the long component present in the decay to the total light output, were measured. Imax is the intercept at time t = 0 in the plots shown in figures II.1 and 2. The value of R can readily be shown to be given by $R = \frac{I_1\tau}{I_1\tau+I_2t}$ where τ and t are the components present in the decay and I_1 and I_2 are the intercepts shown in figure II2.

II.2(c). Efficiency.

Since all our results could be normalised to the same electronic gain, we have been able to estimate the relative voltage pulse height/unit energy (or efficiency) for the different particles. Two efficiencies were measured: $^{\infty}\xi$ is the efficiency for infinite time based on the assumption that there is no contribution to the light output from components longer than the longest component measured in each case and $^{x}\xi$ is defined as the efficiency for light emitted in the time range up to x μ.sec. The peak height of the voltage pulse was smaller in magnitude than theoretically expected due to the leakage time constant mentioned earlier. However the correction for this effect was of the order of 3% of the peak height and lay within the

experimental error in measuring the peak height ($\pm$ 10%). For this reason the correction has not been applied.

II.3. Results.

Values of the long and short components, t and τ respectively, of the decay time of the luminescence produced by alpha particles, protons and electrons in the crystals studied are shown in tables II.1 to 4. The total analysis time required to obtain these tables was of the order of 200 hours. The subscripts a, p and e are used to denote the type of particle producing the luminescence. In these tables are given the values of ${}^{a}\varepsilon$ and ${}^{x}\varepsilon$ defined earlier. The same subscripts are used to distinguish the different particles. Derived values of I_{max}/E and of R, defined earlier, are also given.

The results given in table II.5 are those presented by Storey et al. (loc.cit.) for CsI(Tl). Our measurements of the values of $\tau_{\alpha,p,e}$ and $t_{a,p,e}$ agreed with their values. For completeness, we have included their complete results.

Table II.6 gives the values of τ'_{α} and τ'_{e} as defined earlier. The values of $\frac{\tau'_e}{\tau'_\alpha}$ and $\frac{\tau_e}{\tau_\alpha}$ are also shown.

It is seen from these tables that in all cases except LiI(Eu) two components are found in the decay.

Table II.1.

LiI(Eu)

Particle	Electron	Proton	Proton	Proton	Proton	Alpha
Energy (MeV)	0.66	15	10	6	3	3.5
Short component (μ.sec)	All gave value τ = 0.94 ± 0.06 μ.sec.					
Long component	No long component detected					
$^{\infty}\varepsilon$	1±0.05	0.87±0.03	-	-	-	-

Table II.2.

NaI(Tl).

Particle	Electron	Proton	Proton
Energy (MeV)	0.66	2.5	14.9
Short component τ (μ.sec)	0.23±0.01	0.21±0.01	0.23±0.01
Long component t (μ.sec)	1.5±0.08	1.5±0.08	1.5±0.08
$^{5}\varepsilon$	1±0.05	-	1.15±0.06
I_{max}/E	1±0.1	-	1±0.1
R	0.43	0.4	0.38

Table II.3.

KI(Tl).

Particle	Electron	Proton	Proton	Proton	Proton	Alpha
Energy (MeV)	0.66	14.02	9.7	6.8	3.14	3.4
Short compt. (μ.sec)	0.24 ± 0.01	0.2 ± 0.02	0.22±0.02	0.2±0.01	0.2±0.008	0.21±0.01
Long compt. t (μ.sec)	2.5 ± 0.1	1.9 ± 0.09	1.6±0.1	1.56±0.08	1.6±0.08	1.76±0.09
0.5ε	1 ± 0.05	1.31 ± 0.06	1.27±0.06	1.28±0.06	1.27±0.06	1.07±0.05
$^{6}\varepsilon$	1 ± 0.05	1.29± 0.08	1.28±0.07	1.22±0.06	1.53±0.08	1.06±0.08
I_{max}/E	1 ± 0.1	1.54 ± 0.15	1.93±0.19	1.3 ±0.13	1.53±0.15	1.53±0.15
R	0.44	0.39	0.35	0.38	0.39	0.3

Table II.6.

Values of $\tau'_{\alpha,e}$ the average lifetime after 4 lifetimes of electron short component.

Crystal	τ'_α	τ'_e	$\frac{\tau'_e}{\tau'_\alpha}$	$\frac{\tau_e}{\tau_\alpha}$
LiI(Eu)	0.9	0.9	1	1
NaI(Tl)	0.335 *	0.35	1.04 *	1.09
KI(Tl)	0.43	0.48	1.13	1.14
CsBr(Tl)	2.86	3.52	1.23	1.12
CsI(Tl)	1.17	1.6	1.37	1.68

* The value quoted here as τ'_α is in fact the value for a low energy proton. Hence τ'_e/τ'_α will be slightly greater than 1.04.

In the results given for NaI(Tl), table II.2, no measurements were made with alpha particles since they were stopped in the window in the air-tight aluminium container in which the crystal was mounted. Since the exact thickness of the aluminium window was not known, $^{\infty}\varepsilon_p$ has not been measured for low energy protons. Our value of γ_e is in good agreement with the result of Eby and Jentschke (1954) who quote $\gamma_e = 0.25$ μ.sec. They however did not find any difference in the values of $\gamma_{\alpha,p,e}$. Nicholson and Snelling (1955) find a value of 0.34 μ.sec. for the decay time for electrons. This fits our average value given in table II6.

For KI(Tl) our value of $^{\infty}\varepsilon_p \,|\, ^{\infty}\varepsilon_\alpha = 1.29 \pm 5\%$, where $^{\infty}\varepsilon_p$ is in this case the value of efficiency for 14.02 MeV protons. This value is in agreement with the result of Franzen et al. (1950) who find a value of $1.12 \pm 10\%$ where $^{\infty}\varepsilon_p$ is in this case for 16 MeV protons. The value of $^{\infty}\varepsilon_p \,|\, ^{\infty}\varepsilon_\alpha$ obtained by Kienle and Segel (1959) of $1.42 \pm 5\%$ is in good agreement with our value of 1.53 ± 0.08, where $^{\infty}\varepsilon_p$ is in this case the value of efficiency for 3 MeV protons. Our value of $^{\infty}\varepsilon_\alpha \,|\, ^{\infty}\varepsilon_e = 1.07 \pm 10\%$ is in good agreement with the value of $0.92 \pm 5\%$ found by Kienle and Segel (loc.cit.), and with the value 0.96 found by Franzen et al. (loc.cit.).

$^{\infty}\varepsilon$ was not measured for lower energy protons or for alpha particles in LiI(Eu) since to prevent the crystal being exposed to air it was completely enclosed in silicone grease. The layer of silicone grease was thin enough to have a negligible effect on the energy of 15 MeV protons. Schenk and Neiler (1954) find that in LiI(Eu), $^{\infty}\varepsilon$ first increases and then decreases with increasing ionization density ρ due to the exciting particle. Our results do not agree with this. The difference may be due to different europium concentrations. Nicholson and Snelling (1955) find τ = 1.4 μ.sec. for their crystal LiI(0.03% Eu). Our results in a standard Harshaw (CsI(Tl 0.1%)) crystal substantiate those of Storey et al. (loc.cit.) given in table II.5. These results are in agreement with those of Fleury et al. (1960) but are in disagreement with those of Owen (1960).

No previous measurements have been made in CsBr(Tl) except by Bitman et al. (1952) who find the qualitative result that in this crystal and in CsI(Tl) a long component is present in the decay.

For CsBr(pure), the pulse heights produced by electrons and alpha particles were only slightly greater than the luminescence of the glass of the photomultiplier. $\tau'\rho$ is estimated as 10.5 ± 1 μ.sec. for protons of energy 14.9 MeV. No luminescence was produced in a pure NaCl crystal.

II.4. Summary of Results.

These results can be summarised as follows.

The results are consistent with the picture that the detailed shape of the decay of the luminescence depends on the average ionization density ρ due to the particle producing the luminescence.

In addition we find:-

(1) $\tau_\alpha \leq \tau_e$

(2) τ increases for all particles as Z of the cation increases.

(3) t_e for CsBr(Tl) and KI(Tl) $>$ $t_{\alpha,p}$

(4) ${}^{\infty}\varepsilon_\alpha < {}^{\infty}\varepsilon_e$

(5) In CsI(Tl) and KI(Tl), ${}^{\infty}\varepsilon$ first increases and then decreases with the ionization density ρ of the ionizing particle. In CsBr(Tl) the results obtained are too similar to say if this is the case or not.

(6) τ_e/τ_α increases as Z of the cation increases.

Any model which is proposed to explain the luminescent properties of the alkali halides must account for these facts.

In table II.6 the ratio τ'_e/τ'_α can be seen to increase as Z of the crystal increases. From the practical point of view it would be of interest therefore to study crystals of very large Z; of the crystals studied it is seen that none has properties more suitable than CsI(Tl) for use in a

technique similar to that described in Part I.

II.5. The Luminescence of CsI(Tl).

We now consider several measurements which were made on the luminescence of CsI(Tl). These have a bearing on the validity of the results presented in the above section.

It was recognised that the result of Storey et al. (loc.cit.), that the decay time of the luminescence produced in CsI(Tl) by ionizing particles was apparently dependent on the ionization density produced by the particle, might not have been an intrinsic property of the luminescence but have been caused by external sources.

Firstly, the measured values of the decay times might well be influenced by the type of photomultiplier used in making the measurements. For this reason measurements were made using an R.C.A.6810, a Du Mont 6292 and an E.M.I.5311 photomultiplier. In each case the same CsI(Tl) crystal was used in making the measurements. The decay times of the luminescence produced by alpha particles and electrons, were measured in the usual way and it was found that for any one photomultiplier the effect did indeed exist although there were slight differences in the decay time of the luminescence for any particular particle, as measured by different photomultipliers. These discrepancies, however, fell within the limits of experimental error.

It can be concluded that all the commonly available glass window photomultipliers give the same results and that the difference in the values of the decay times of the luminescence produced by different particles is not caused by the type of photomultiplier used.

The effect might have been arising in the following manner. The path length of the light passing through the crystal to the photocathode of the photomultiplier is greater in the case of alpha particles than for electrons, and this difference could readily give rise to differential absorption of initially longer lifetime components in the light pulses of the alpha particles. This, in turn, would lead to a shorter measured value of the lifetime of the emission produced by alpha particles. This possibility can be rejected, however, since measurements made on crystals of varying thickness were identical provided the crystal was sufficiently thick to stop the exciting particle.

Further evidence for the rejection of a differential absorption process was afforded by mounting a CsI(Tl) crystal in a reflecting cavity a few centimetres from the face of the photomultiplier and allowing alpha particles to enter on the surface nearest the photocathode. In this way the path of the light emitted after alpha particle excitation was reduced to a minimum. The value of τ_α obtained from

this experiment was the same as when the crystal was excited by alpha particles entering the surface furthest from the photocathode.

Finally, fairly strong evidence, that the decay time of the luminescence produced by a particle does depend on the average ionization density produced by the particle, was obtained from measurements of the decay time of the luminescence produced by 14 MeV protons in a CsI(Tl) crystal sufficiently thick to stop the protons, and in a CsI(Tl) crystal in which the protons lost only part of their energy (≈ 4 MeV). It was found that the decay time of the luminescence was greater for the thin crystal in which the average ionization density produced by the protons is smaller. This is in accordance with the expected behaviour of τ with ρ , the average ionization density.

From these measurements it can be concluded, that the result presented in the previous section, that the decay time of the luminescence produced by a particle in alkali halide crystals depends on the ionization density of the particle, is an intrinsic property of the mechanism of the luminescence and not produced by artificial means.

Part II.

Chapter III.

The Luminescence of CsI(Tl) and CsBr(Tl) as a Function of Temperature and the Luminescence of CsI(Tl) as a Function of Thallium Concentration.

The results presented in the previous chapter establish the fact that the decay time of the luminescence produced in alkali halide crystals by ionizing particles is dependent on the average ionization density produced by the exciting particle. In addition, they show that of the commonly available inorganic crystals, CsI(Tl) and CsBr(Tl) are most suitable for use with the technique discussed in Part I of this thesis. The measurements presented in this chapter were made in the hope that a more favourable particle discrimination might be obtained by maintaining the crystal at temperatures other than room temperature, or by altering the thallium concentration. The behaviour of the decay times and efficiencies of the luminescence as a function of temperature, or thallium concentration, is also important when considering the mechanism of the luminescence.

III.1. The Luminescence of CsI(Tl) and CsBr(Tl) as a Function of Temperature.

III.1(a). Experimental Technique.

In order to vary the crystal temperature it was necessary to design a piece of apparatus which would incorporate the following features:-

(1) A means whereby the crystal temperature could be varied without varying the temperature of the photocathode an effect which is known to influence the gain of a photomultiplier. This necessitates the use of some kind of chamber, with good reflecting walls, in which the crystal can be housed, the chamber then being mounted on the photomultiplier.

(11) The chamber must be vacuum tight otherwise on allowing the cold crystal to warm, condensation of water would occur on its surface, prevent the entry of alpha particles, affect the response of the crystal and impair the resolution. In addition at low temperatures the crystal would be surrounded by a coating of ice which would again reduce the energy of alpha particles and spoil the efficiency measurements.

Furthermore, the presence of cold air inside the chamber would tend to produce cooling at the photocathode.

(111) A means of measuring the crystal temperature. For this purpose it was decided to use a thermocouple.

The assembly shown in figure III.1 was found to be satisfactory.

It consists of a cylinder of stainless steel (1/32" thick x 2" in diameter) lined with aluminium foil reflectors.

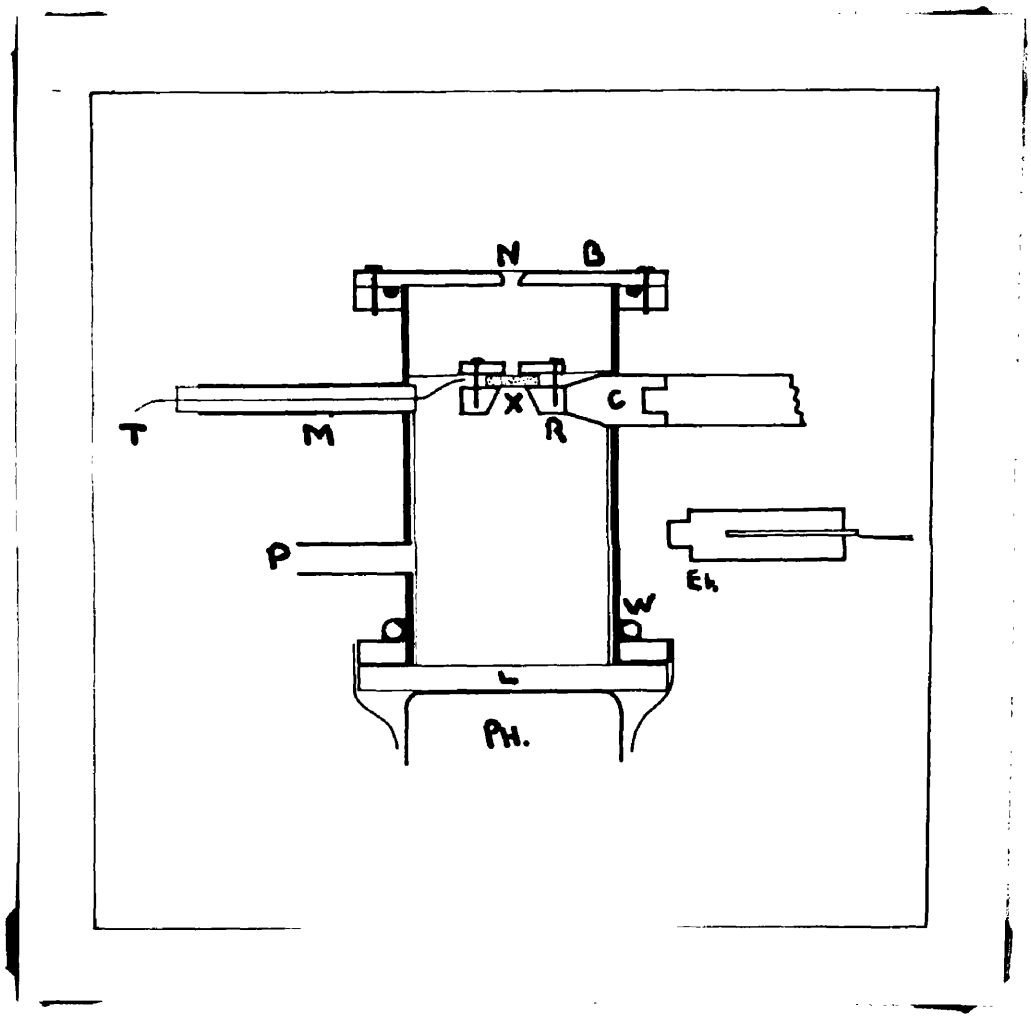


Figure III.1. Crystal Assembly.

The top plate of the cylinder (B) was detachable and made of brass in the centre of which is a thin nickel window (N) (2 mg./cm^2). The base of the cylinder was closed with a disc of lucite (L) which was optically coupled to an R.C.A.6810 photomultiplier with silicone grease. A thin aluminium foil (0.2 mg./cm^2) backed the crystal (X) which was clamped between two copper rings (R) both of which had aluminised surfaces. The lower ring was attached to a copper rod (C) which protruded through the wall of the vessel where it could be cooled with liquid nitrogen or heated with an electrical element (El) as required. A pumping port (P) is incorporated in the cylinder by means of which it can be evacuated. A thermocouple (T) was used to measure the crystal temperature. The thermocouple wires were led out of the chamber by two short pieces of capillary glass extending through the outer wall and supported by metal collars (M), vacuum seals being made using araldite. This prevented short circuiting across the stainless steel wall. These seals were situated fairly far from the crystal, to prevent cracking whilst cooling is in progress. Finally water cooling pipes (W) ensured that the temperature of the photocathode (Ph) remained constant.

All heating and cooling was carried out slowly and no hysteresis effect was observed. The range of temperature obtainable was -160°C to +160°C.

The CsI(Tl) and CsBr(Tl) crystals used in the measurements were obtained from the Harshaw Chemical Company, the thallium concentration in each case being 0.1% molar.

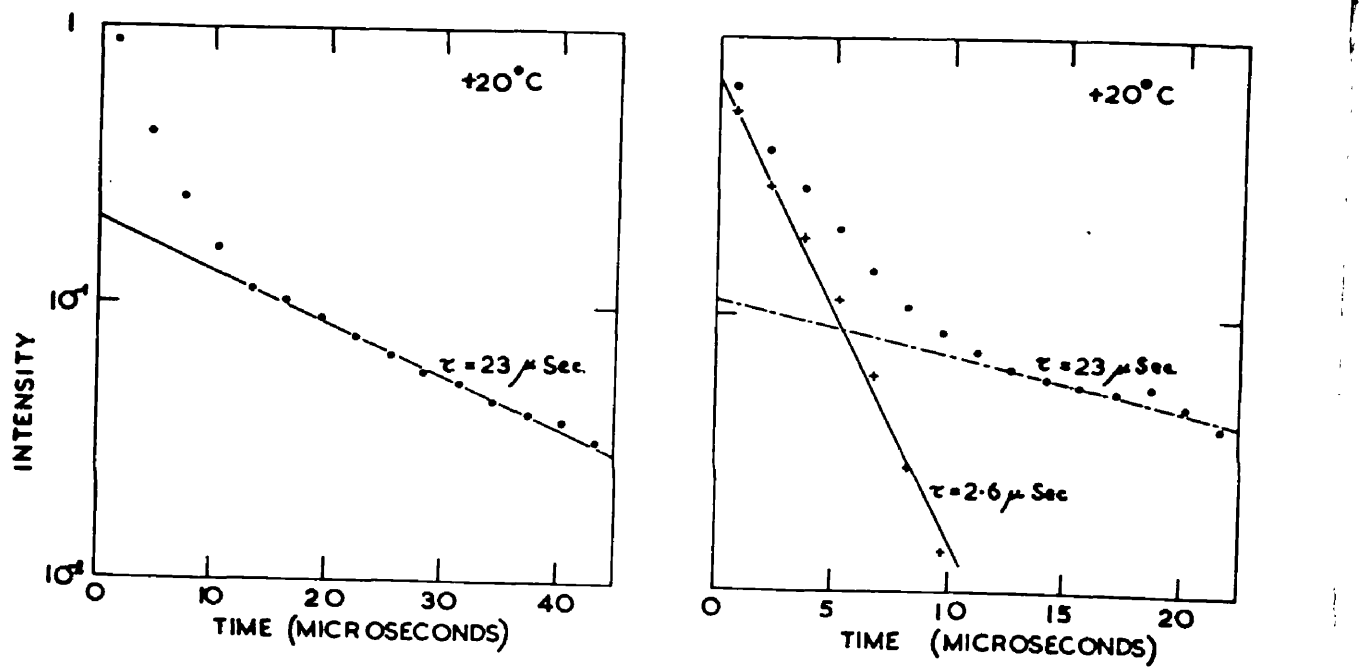
In these measurements, the particles producing high and low ionization density were respectively, alpha particles of energy 5.3 MeV from ^{210}Po and protons of energy 14 MeV from the reaction $He^3(d,p)He^4$.

The current pulses from the photomultiplier were integrated with a leakage time constant of 205 μ.sec. These 'voltage' pulses were displayed on a Tektronix type 541 oscilloscope and photographed at fixed temperature intervals.

III.1(b). Analysis.

The decay of the luminescence was obtained by the method described in Chapter II. The analysis of the decay of the luminescence of CsBr(Tl) at +20°C irradiated with protons of 14 MeV is shown in figure III.2. From figure III.2a, it appears that an exponential component of time constant τ = 23 μ.sec. is present. Having estimated the lifetime and contribution of this component from figure III.2(a), it is subtracted from a pulse on a shorter time scale, figure III.2(b). The resulting points are in agreement with a pure exponential of time constant τ = 2.6 μ.sec.

In general, at any one temperature, the decay of the luminescence of CsI(Tl) and CsBr(Tl) may be described by



(a) (b)

Figure III.2. The Decay Curves of the Luminescence produced by 14 MeV protons in CsBr(Tl) at +20°C. A comparison of this figure with figure II.2 illustrates the accuracy obtainable by this method of analysis.

the sum of a fast and a slow exponential component. Figure III.3 shows one of the exceptional cases where three exponential components appear to be present in the luminescence produced by fast proton irradiation of CsBr(Tl) at +120°C.

In CsI(Tl) the contribution from the slow exponential component is small over most of the temperature range. Accordingly, the pulse has been analysed by an alternative method which assumes that the decay of the luminescence is a pure exponential at times short compared to the lifetime of the slow exponential component. Values derived by this method are denoted by τ'. The subscripts α and p denote excitation of the crystal by alpha particles and protons respectively.

III.1(c). Results.

Plotting the logarithm of the decay time components against the reciprocal of the absolute temperature, figures III4a,b and III.5a,b, it is seen that the components obtained by the above analysis are not distributed randomly, but, in general, follow exponential laws. In order to discriminate between the different components present in the decay, the components have been numbered according to the line on which they lie.

The variation with temperature of the pulse height per unit energy or luminescent efficiencies $^{\infty}\varepsilon_{\alpha,p}$ is

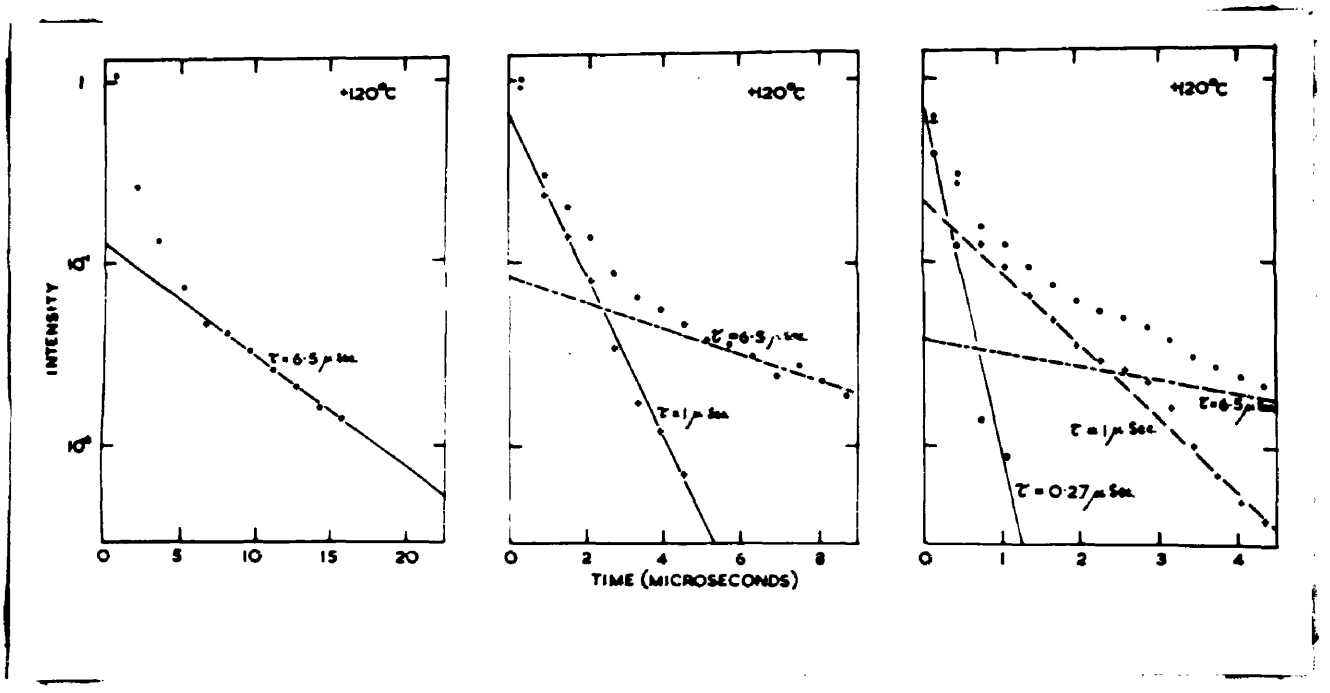


Figure III.3. The Decay Curves of the Luminescence produced by 14 MeV protons in CsBr(Tl) at +120°C.

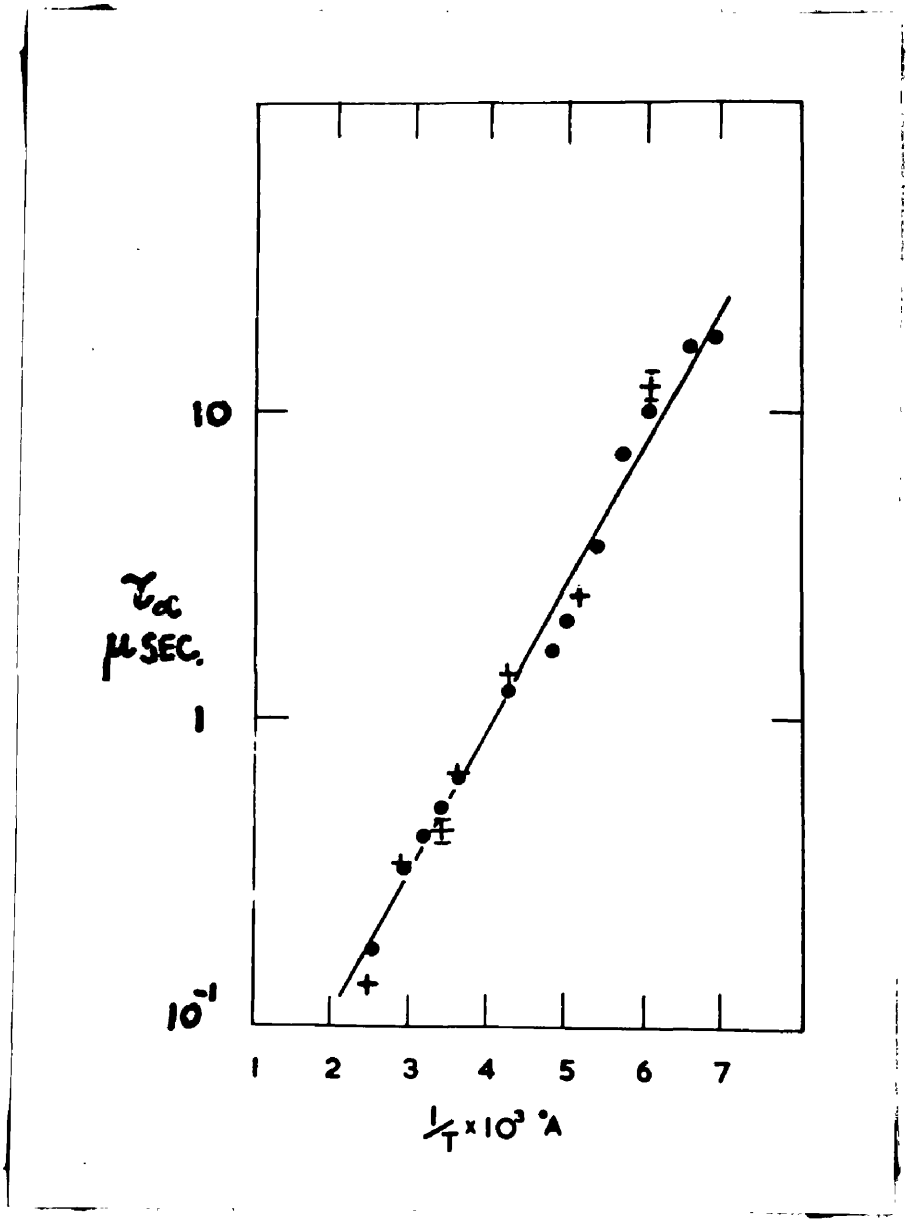


Figure III.4(a). Alpha particle excitation of CsI(Tl). Decay time component $\tau_{1\alpha}$ as a function of $\frac{1}{T}$

+ $\tau_{1\alpha}$ values; ● $\tau'_{1\alpha}$ values.

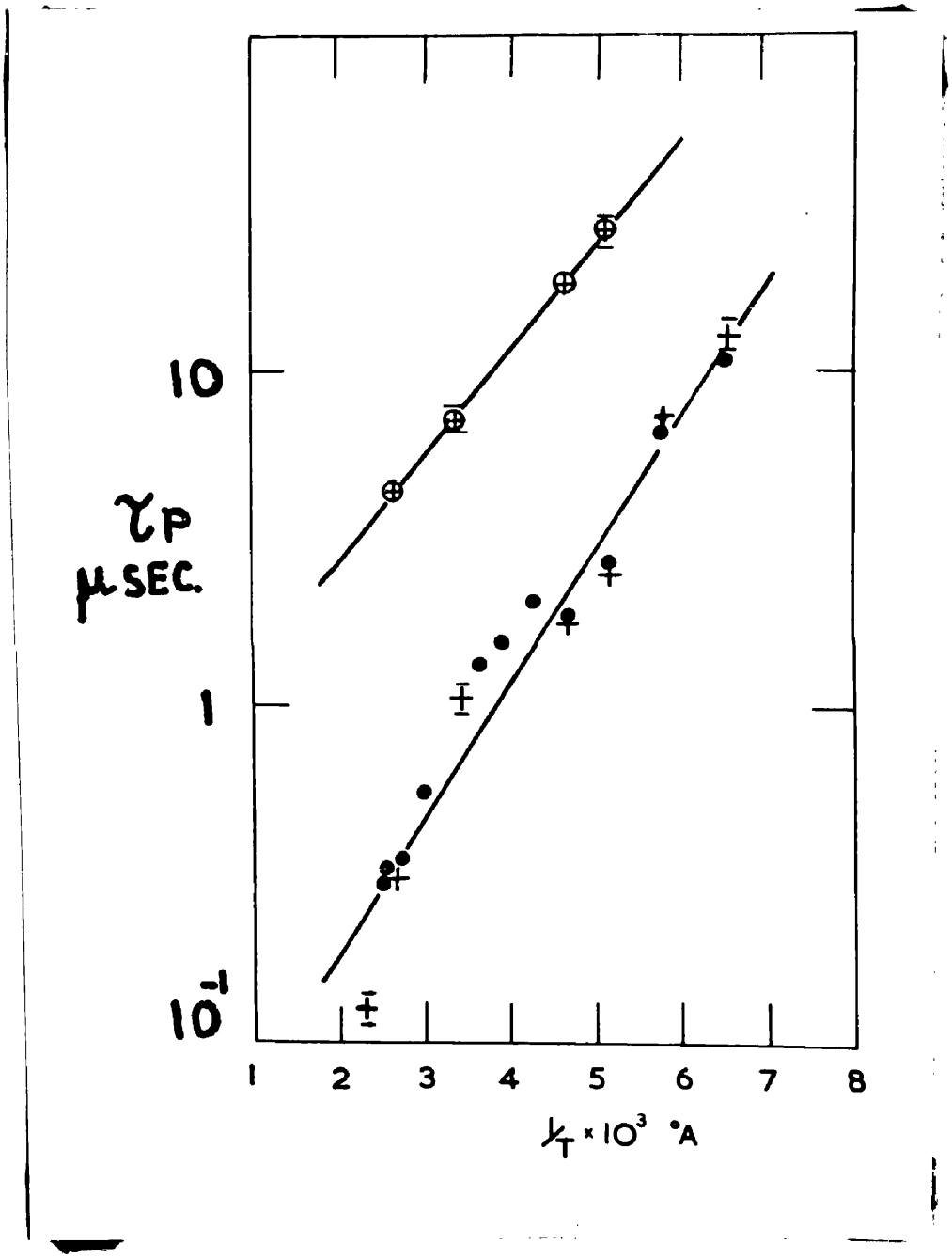


Figure III.4(b). Proton excitation of CsI(Tl)
Decay time components γ_{1p} and γ_{2p} as a function of $\frac{1}{T}$

+ γ_{1p} values, ● γ'_{1p} values,

⊕ γ_{2p} values.

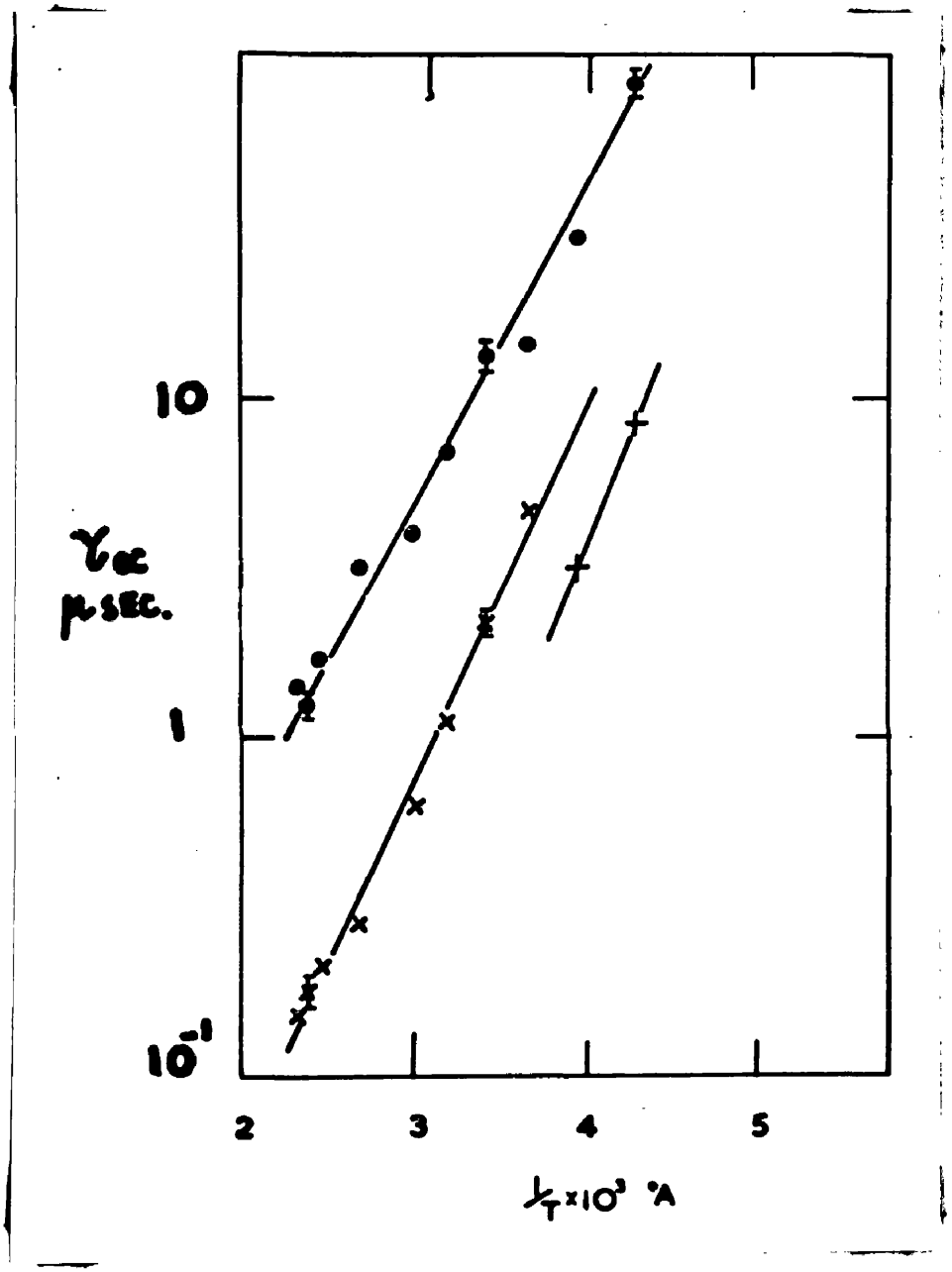


Figure III.5(a). Alpha particle excitation of CsBr(Tl). Decay time components $\gamma_{i\alpha}$ as a function of $\frac{1}{T}$

+ $\gamma_{1\alpha}$ values, X $\gamma_{2\alpha}$ values,

● $\gamma_{3\alpha}$ values.

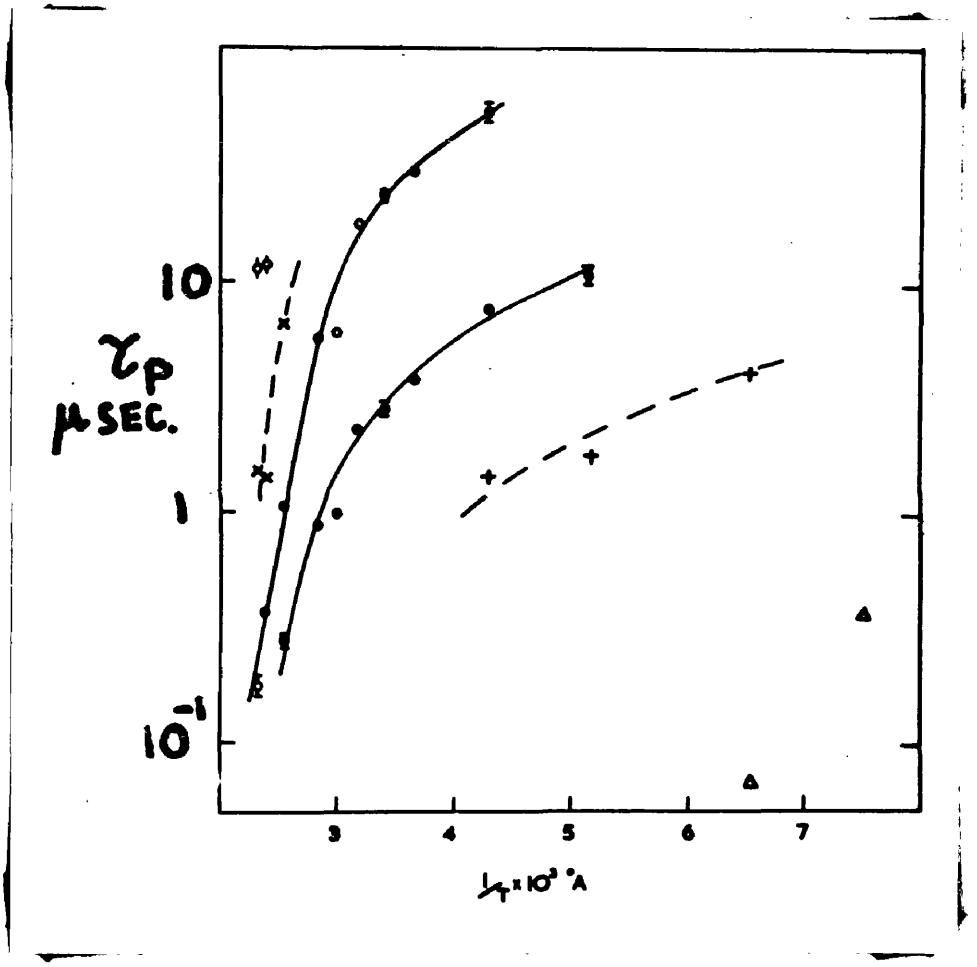


Figure III.5(b). Proton excitation of CsBr(Tl). Decay time components τ_{ip} as a function of $\frac{1}{T}$.

Δ τ_{1p} values, + τ_{2p} values,

● τ_{3p} values, o τ_{4p} values,

x τ_{5p} values, ϕ τ_{6p} values.

shown in figures III.6 and III.7. In these efficiency measurements it was assumed that all the components contributing to the light output were observed. The results shown have been corrected for the effect of the electronic differentiation time constant on the pulses.

Tables III.1a,b and III.2a,b give the values of R_1, R_2 etc. the ratios of the light present in components 1,2, etc. to the total light output observed.

In all cases the decay time components become shorter as the temperature of the crystal increases.

From figures III.4a and III.5a for the alpha particle excitation of the crystals, it appears that the decay time components in these cases have an exponential dependence on $\frac{1}{T}$, where T is the absolute temperature. It is customary, Bonanomi and Rossel, 1952; Enz and Rossel, 1958 to derive from these plots a value for the activation energies of the electron traps associated with the luminescence. The assumption is made that the probability of a luminescent transition is proportional to $\exp(+\frac{E}{kT})$ where E is the electron trap activation energy and k is Boltzmann's constant. For CsI(Tl) we obtain $E_{1\alpha} = 0.092 \pm 0.01$ e.v., while CsBr(Tl) yields values $E_{2\alpha} = 0.22 \pm 0.02$ e.v. and $E_{3\alpha} = 0.19 \pm 0.02$ e.v. Although the decay time components for proton excitation of CsI(Tl) τ_{1p} and τ_{2p} show an approximately exponential dependence on $\frac{1}{T}$ yielding electron

Table III.1(a)

Values of R for alpha particle excitation of CsI(Tl).

Temperature	-80°C	-40°C	0°C	+20°C	+80°C	+146°C
R_1	1	1	0.86	0.75	1	1
R_2	-	-	0.14	0.25	-	-

Table III.1(b)

Values of R for Proton excitation of CsI(Tl).

Temperature	-120°C	-100°C	-80°C	-60°C	+20°C	+100°C	+155.5°C
R_1	1	1	0.32	0.39	0.55	0.93	1
R_2	-	-	0.68	0.61	0.45	0.07	-

Table III.2(a).

Values of R for alpha particle excitation of CsBr(Tl).

Temperature	-40°C	-20°C	0°C	+20°C	+40°C	+60°C	+100°C	+130°C	+147°
R_1	0.19	0.16	-	-	-	-	-	-	-
R_2	-	-	0.23	0.72	0.48	0.56	0.58	0.69	0.74
R_3	0.81	0.84	0.77	0.28	0.52	0.44	0.42	0.31	0.26

Table III.2(b).

Values of R for proton excitation of CsBr(Tl).

Temperature	-120°C	-80°C	-40°C	0°C	+20°C	+40°C	+60°C	+80°C	+120°C	+147°C	+156
R_1	0.26	-	-	-	-	-	-	-	-	-	-
R_2	0.74	0.51	0.18	-	-	-	-	-	-	-	-
R_3	-	0.49	0.16	0.37	0.42	0.47	0.42	0.37	0.27	-	-
R_4	-	-	0.66	0.63	0.58	0.53	0.58	0.63	0.35	0.39	0.18
R_5	-	-	-	-	-	-	-	-	0.38	0.37	0.46
R_6	-	-	-	-	-	-	-	-	-	0.24	0.37

trap activation energies of E_{1p} = 0.079 ± 0.01 e.v. and E_{2p} = 0.062 ± 0.02 e.v., there is no evidence of this exponential behaviour in the proton excitation of CsBr(Tl).

The temperature dependence of the ratio $\gamma_{1p}/\gamma_{1\alpha}$ for CsI(Tl) is shown in figure III.8. The ratio decreases as the temperature of the crystal is lowered, until at approximately -80°C, $\gamma_{1p}=\gamma_{1\alpha}$. At lower temperatures $\gamma_{1\alpha}>\gamma_{1p}$

From the ratios R_1, R_2 etc., it can be seen that the amount of light emitted in any particular component relative to the total light output, alters with temperature.

In figures III.6 and 7, it is seen that the luminescent efficiencies of CsI(Tl) and CsBr(Tl) have a maximum value in the neighbourhood of room temperature. An interesting feature is the temperature range -70°C to -140°C in CsI(Tl) where the luminescent efficiency for alpha particles excitation is greater than that for proton excitation. This effect is also observed to a lesser degree in CsBr(Tl).

III.2. <u>The Luminescence of CsI(Tl) as a Function of Thallium Concentration</u>.

III.2(a). <u>Experimental Technique</u>.

Measurements of the decay times of the luminescence produced by alpha particles, protons of varying energies and electrons have been made on a standard Harshaw CsI(Tl) crystal, a CsI crystal with very low thallium concentration,

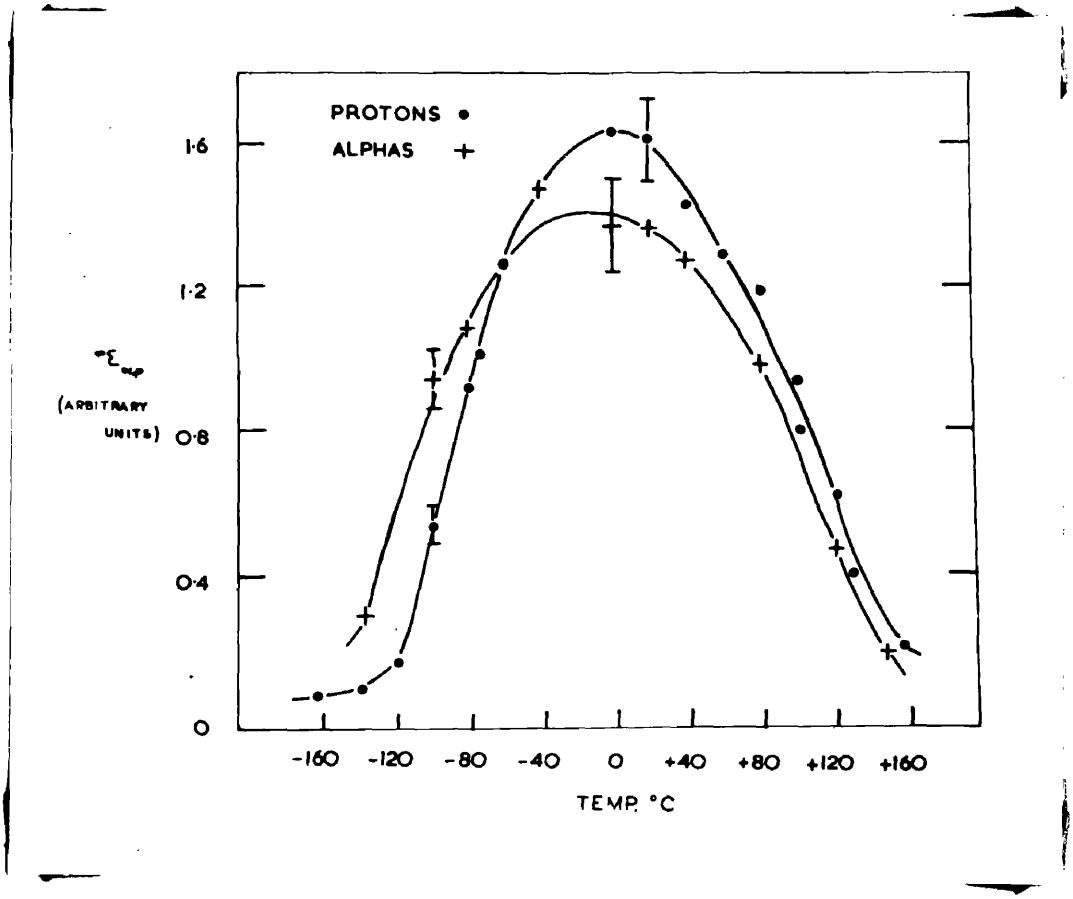


Figure III.6. The Luminescent efficiencies, $^{\infty}\varepsilon_{\alpha,p}$ in CsI(Tl) as a function of temperature.

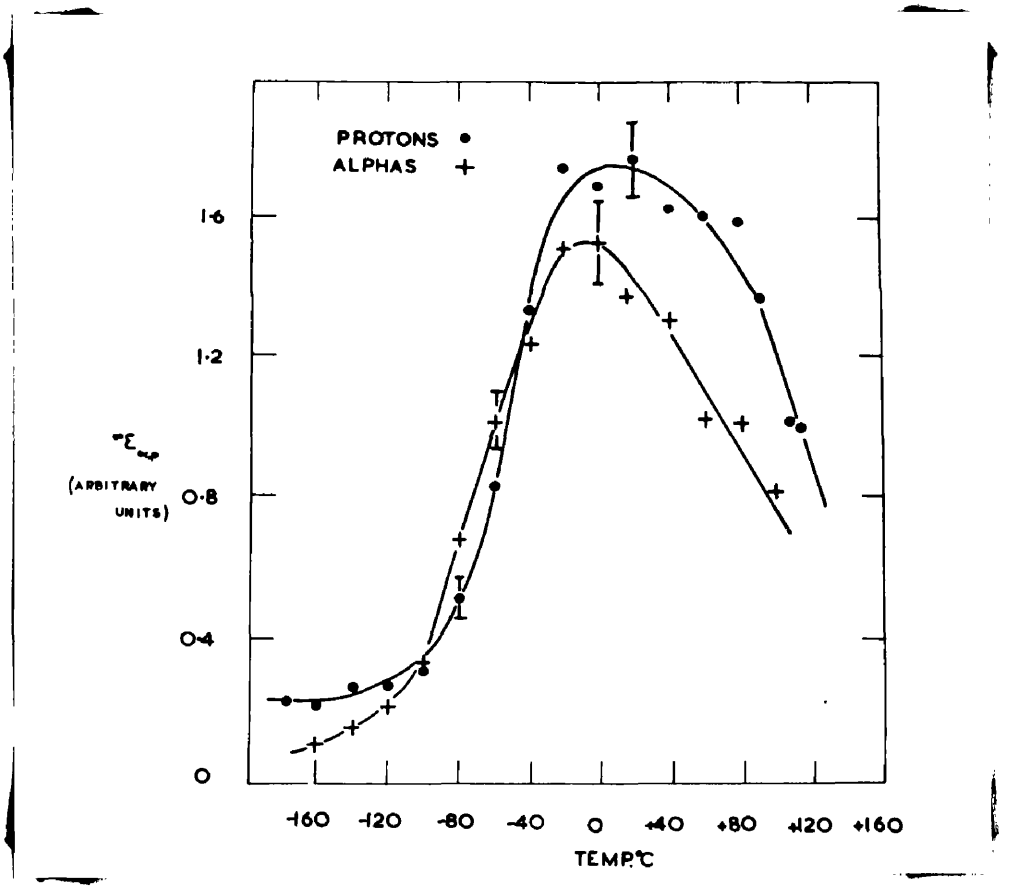


Figure III.7. The luminescent efficiencies, $^{\infty}\varepsilon_{\alpha,\rho}$ in CsBr(Tl) as a function of temperature.

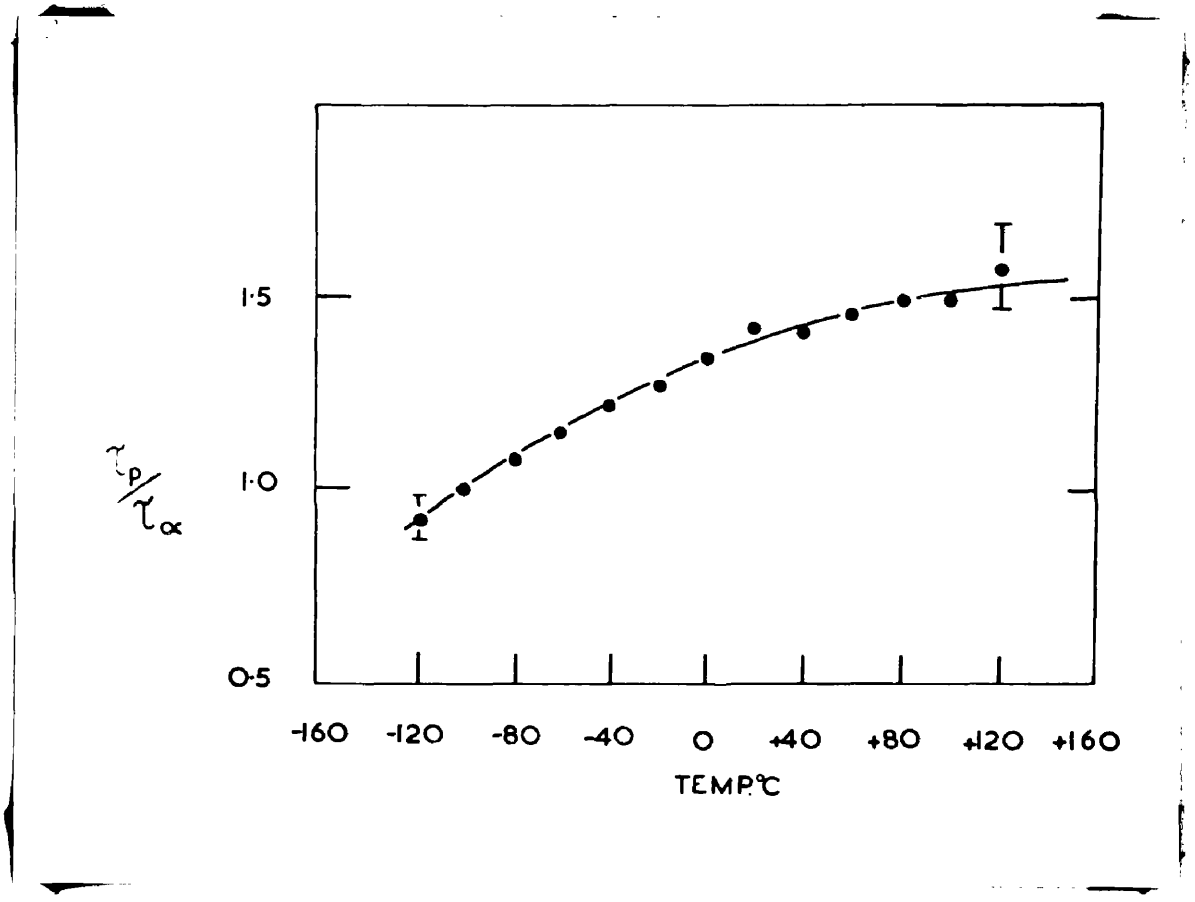


Figure III.8. Variation of the ratio $\gamma_{1p}/\gamma_{1\alpha}$ with temperature in CsI(Tl).

and a CsI crystal with high thallium concentration. The CsI crystals of high and low thallium concentrations (3% and 0.009% respectively) were obtained from Aberdeen University. The bulk crystals were grown in a vacuum furnace similar to that described by Stockberger (1949). The crystals used were cut from the interior of larger crystals and were cylinders 1/4 inch in diameter by 1/4 inch long. The thallium concentrations were not determined directly, the figures quoted here refer to the values obtained using the known weight of thallous iodide added to the crucible and the final weight of the crystal taken from the furnace. The fraction of thallium lost in the growth is assumed to be constant and not excessive.

The experimental technique was exactly similar to that described in Chapter II.

III.2(b). Results.

Table III.3 gives the values of $\tau'_{\alpha,p,e}$ and $^{\infty}\varepsilon_{\alpha,p,e}$ for the three crystals. $\tau'_{\alpha,p,e}$ was measured over the first 4 μ.sec. of the voltage pulse from these crystals, assuming the decay to be a pure exponential. τ' is the time taken for the voltage pulse to rise to $1-\frac{1}{e}$ of its height after 4 μ.sec. $^{\infty}\varepsilon_{\alpha,p,e}$ in the Harshaw crystal (0.1% Tl) was not measured since the geometry of this

Table III.3.

Variation of $\tau_{\alpha,p,e}$ and $^{\infty}\varepsilon_{\alpha,p,e}$ in CsI(Tl) with varying Thallium Concentration.

Thallium concentration	τ'_{α} (μ.sec)	τ'_{p} μ.sec. 3.14 MeV	15 MeV	τ'_{e} (μ.sec)	$^{\infty}\varepsilon_{\alpha}$	$^{\infty}\varepsilon_{p}$ 3.14 MeV	15 MeV	$^{\infty}\varepsilon_{e}$
0.009%	1.55±0.05	1.77±0.06	1.5±0.05	1.58±0.02	0.036	0.08	0.6	0.746
0.1%	1.17±0.05	-	-	1.6 ±0.02	-	-	-	-
3%	1.07±0.05	1.23±0.06	1.5±0.05	1.58±0.02	0.746	0.9	1.07	1

crystal differed from that of the other two. In figures III.9 and III.10 the measurements of $\gamma'_{\alpha,e}$ and $^{\infty}\varepsilon_{\alpha,e}$ are compared with those of Jones and Ward (1960). In these figures, the values of γ'_e and $^{\infty}\varepsilon_e$ have been normalised to unity in the standard Harshaw crystals since this was the manner in which Jones and Ward presented their results. The measurements for alpha particle and electron excitation of the crystals of high and low thallium concentration should be identical since the crystals used were the same, our measurements being made on the crystals supplied by Jones and Ward. This is in fact the case, within experimental error. Our standard Harshaw crystal was different from that of Jones and Ward but the results again substantiate their values. Considering the value of $^{\infty}\varepsilon_{\alpha}$ in the standard Harshaw crystal obtained by Jones and Ward, and in the present measurements, it would appear that Jones and Ward have overestimated the amount of thallium entering the crystals in their growth and that in fact the thallium concentrations in the crystals grown at Aberdeen should be taken to be a factor of approximately ten smaller. Our measurements, in accord with those of Jones and Ward (loc.cit.) indicate that γ'_{α} decreases with increasing thallium concentration and reaches a minimum at approximately the concentration present in the standard Harshaw crystals.

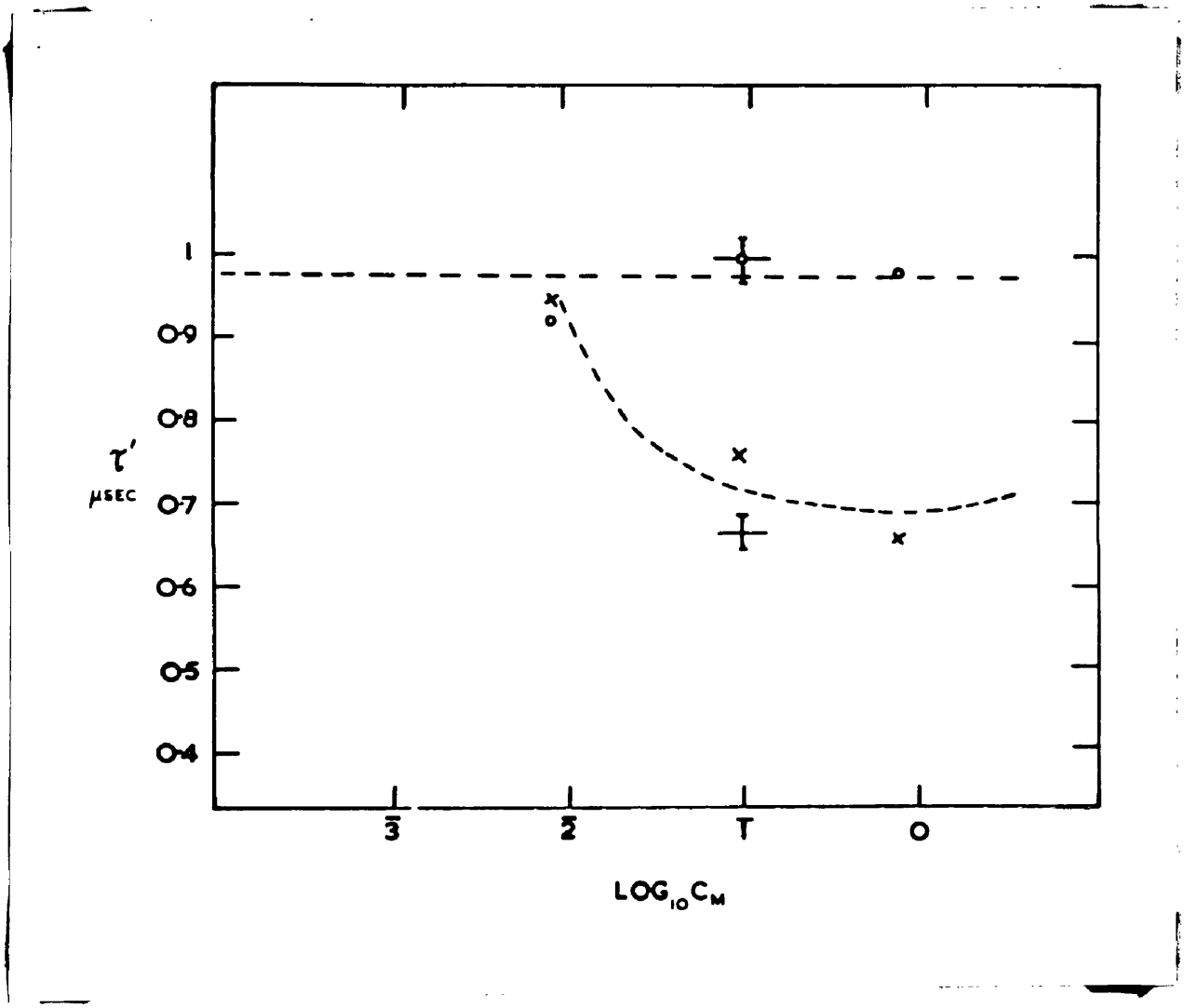


Figure III.9. The Decay times $\tau'_{\alpha,e}$ of the luminescence in CsI(Tl) as a function of thallium concentration. The dashed curves indicate the results obtained by Jones & Ward (1960). The error in all points is ±10%.

o Values of τ'_e , X values of τ'_α

⊕ τ'_e in a standard Harshaw crystal as measured by Jones & Ward

⊕ τ'_α " " " " "

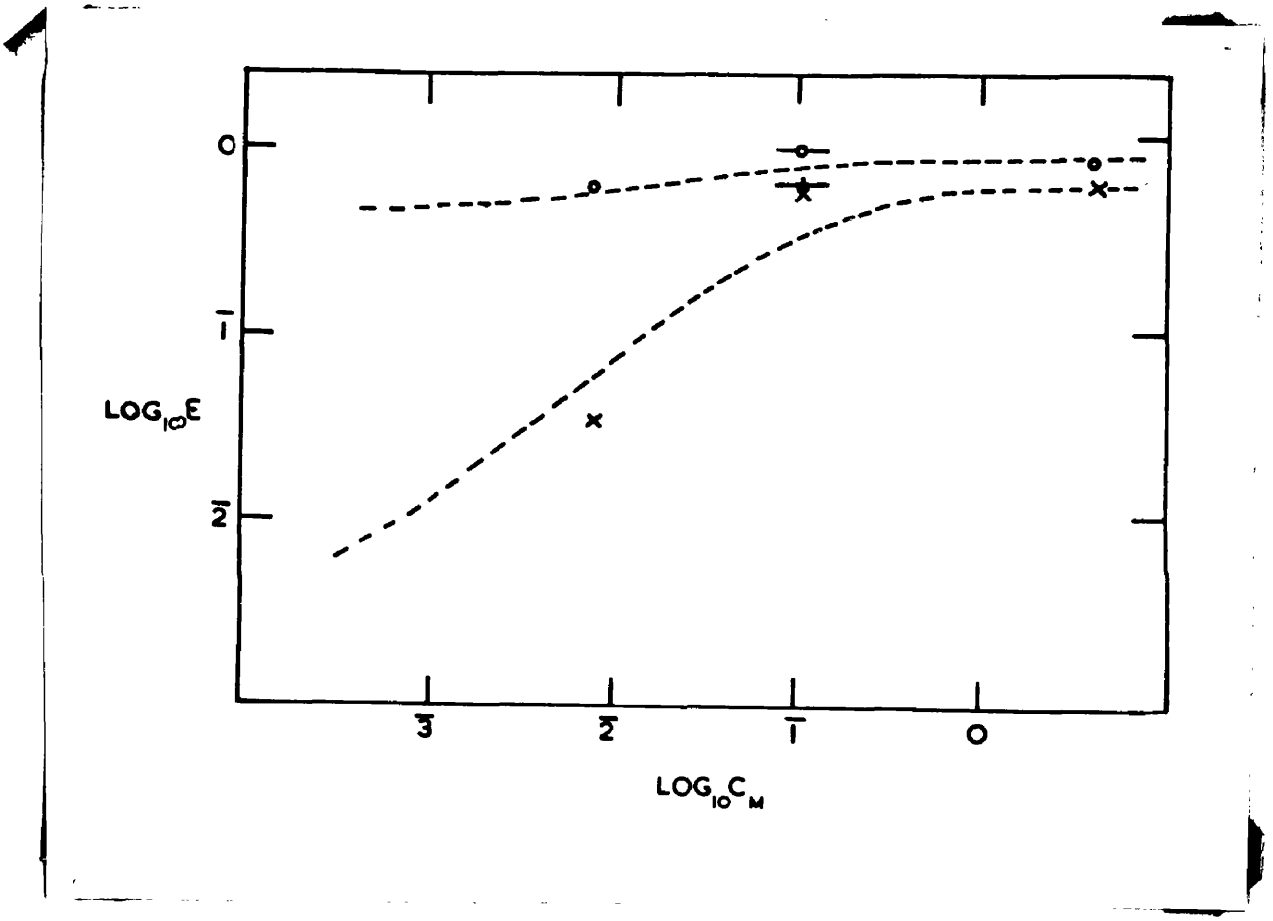


Figure III.10. The luminescent efficiencies ${}^{\infty}\varepsilon_{\alpha,e}$ in CsI(Tl) as a function of thallium concentration. The dashed curves indicate the results obtained by Jones and Ward (1960). The error in all points is $\pm$10%.

o values of ${}^{\infty}\varepsilon_{e}$, x values of ${}^{\infty}\varepsilon_{\alpha}$

-o- value of ${}^{\infty}\varepsilon_{e}$ in a standard Harshaw Crystal as measured by Jones & Ward

-+- value of ${}^{\infty}\varepsilon_{\alpha}$ " " " "

τ'_e on the other hand remain constant with varying thallium concentration.

$^{\infty}\varepsilon_\alpha$ decreases as the thallium concentration ~~increases~~ DECREASES while $^{\infty}\varepsilon_e$ decreases much less markedly.

In fact a study of the values of $\tau'_{\alpha,p,e}$ and $^{\infty}\varepsilon_{\alpha,p,e}$ indicate that the decay time of the luminescence, and the efficiency of the luminescence, produced by a particle in a CsI(Tl) crystal are not governed so much by the thallium concentration present in the crystal, but that the ionization density produced by the particle is the governing factor.

III.3. Conclusions.

From the point of view of particle discrimination in CsI(Tl) and CsBr(Tl), we must conclude that there is no advantage to be gained by maintaining the crystals at temperatures other than room temperature.

For CsI(Tl), this follows from a consideration of the behaviour of the ratio $\tau_{1p}/\tau_{1\alpha}$ shown in figure III.8. At temperatures above room temperature, although this ratio increases slightly, the efficiencies $^{\infty}\varepsilon_{\alpha,\rho}$ show a marked decrease. Below room temperature the efficiencies again decrease while the decay times become much longer. This combination of low efficiency and long decay times rules out the use of the scintillator at these temperatures in

the particle discrimination technique.

In CsBr(Tl) the situation is similar, any possible increase in the difference in the shape of the luminescence produced by different particles is counterbalanced by a marked decrease in efficiency.

In CsI(Tl) a consideration of figure III.9 shows that there is no advantage to be gained in pulse shape discrimination by altering the thallium concentration from that present in standard Harshaw crystals.

Part II.

Chapter IV.

Discussion and Conclusions.

In the previous chapters we have stressed the importance of the measurements of the decay times of the luminescence τ and the luminescent efficiencies ${}^{\infty}\varepsilon$ as a function of ionization density, temperature and thallium concentration, from the point of view of the particle discrimination technique described in Part I of this thesis. These measurements of τ and ${}^{\infty}\varepsilon$ are also important in explaining the mechanism of the luminescence processes.

The measurements confirm the suggestion by Storey et al. (loc.cit.) that the decay time of the luminescence produced in inorganic alkali halide phosphors by ionizing particles is dependent on the ionization densities produced by the particles, the decay time being shorter as ionization density increases. Preliminary measurements by the author in LiF, CaF and BaF, and by Scharmann (1959) in $MgWO_4$, $CaWO_4$, $ZnWO_4$ and $CdWO_4$, also confirm this dependence of τ on ionization density. In the alkali halide crystals the dependence of τ on ionization density becomes more pronounced as the atomic number of the crystal increases. No such regular pattern is observed in Scharmann's results in the tungstate crystals. In the alkali iodides τ increases for all particles as Z of the cation increases,

as can be seen from the measurements in NaI(Tl), KI(Tl) and CsI(Tl). Again no such regular behaviour is observed in the tungstate crystals.

The decay time components of the luminescence in CsBr(Tl) and CsI(Tl), are found to increase as the temperature of the crystal is lowered. (Preliminary results in KI(Tl) confirm this behaviour). For alpha particle excitation of the crystals the decay time components show an approximately exponential dependence on $\frac{1}{T}$, where T is the absolute temperature (Figures III.4(a) and 5(a). Although the decay time components for proton excitation of CsI(Tl) again show an approximately exponential dependence on $\frac{1}{T}$, no such exponential behaviour is found in the proton excitation of CsBr(Tl). In CsI(Tl) the decay time of the luminescence τ'_α shows a marked dependence on thallium concentration. τ'_e on the other hand is apparently independent of thallium concentration (figure III.9). It is possible that the phenomena which account for the variation of decay time τ with ionization density due to the particle producing the luminescence, also account for the dissimilar behaviour of τ with $\frac{1}{T}$, and thallium concentration for the different types of particle.

The dependence of ${}^{\infty}\varepsilon$ on ionization density differs from that of the decay time τ . In the alkali halide

crystals the luminescent efficiency shows a maximum with ionization density, whereas if the dependence on ionization was similar to that of τ , we should expect a monatomic decrease with ionization density. This suggests that perhaps the decay time of the luminescence and the luminescent efficiency are determined by different processes.

The variation of $^{\infty}\varepsilon_{\alpha}$ with thallium concentration is much more marked than is the variation of $^{\infty}\varepsilon_{e,\rho}$ (figure III.10 and Table III.5). Although the variation of $^{\infty}\varepsilon_{\alpha}$ and $^{\infty}\varepsilon_{\rho}$ with temperature is similar, there is a region of temperature where $^{\infty}\varepsilon_{\alpha} > {}^{\infty}\varepsilon_{\rho}$ (figures III.6 and 7)($^{\infty}\varepsilon_{\rho}$ here refers to the luminescent efficiency of 14 MeV protons).

Again it is possible, that the dissimilar behaviour of $^{\infty}\varepsilon$ with thallium concentration, for the different particles and the fact that $^{\infty}\varepsilon_{\alpha} > {}^{\infty}\varepsilon_{\rho}$ at low temperatures, may both be caused by the same phenomena which account for the variation of $^{\infty}\varepsilon$ with ionization density.

Clearly any model which is proposed to explain the luminescent properties of the alkali halides must account for these facts. In such models one can take the view that the decay time components are determined independently of the luminescent efficiencies, or that both τ and $^{\infty}\varepsilon$ should be explicable by the same mechanism.

In the following sections we consider how the theoretical explanations discussed in Chapter I can account for the observed experimental results.

IV.1. Possible Explanations of the Luminescent Properties of Alkali Halide Crystals.

The conventional picture for the process producing the luminescence is one in which electrons produced by the ionizing particle are released thermally from traps emitting radiation or in radiationless transitions. The measured decay times show that these radiationless transitions must increase with ionization density due to the exciting particle, resulting in faster decay times of the luminescence for particles of greater ionization density. This is similar to the explanation offered by Scharmann (loc.cit.) for his results in the tungstate crystals. Due to the high ionization density, quenching of the luminescence occurs at the beginning of the decay. Hence the decay is more rapid as the ion mass is increased. It is suggested that the quenching mechanisms may be of the type suggested by Wright (1953) to explain the luminescence efficiency in organic phosphors. On this picture we would expect the decay times for the luminescence to increase as the temperature is decreased due to the decreased probability of thermal release of electrons. Again we would expect the luminescent efficiency to increase as the temperature is lowered since radiationless

transitions are more probable at high temperatures. Both of these, in fact, are found experimentally to occur. However on this picture we would expect the luminescent efficiency to decrease steadily with ionization density but in fact this is not the case. Nor is it clear why at low temperatures the luminescence efficiency should again begin to decrease.

This behaviour of the luminescence efficiency with temperature is a major difficulty in the model presented by Bonanomi and Rossel, and Enz and Rossel (loc.cit.) discussed in Chapter I. In this model, $\frac{1}{\tau} = K_1 + K_2$, where K_1 and K_2 are the probabilities for radiative and non-radiative transitions in the luminescence centre, respectively. $K_1 = S_1 e^{\frac{E_1}{kT}}$ and $K_2 = S_2 e^{\frac{E_2}{kT}}$ where E_1 and E_2 are the activation energies for the radiative and non-radiative transitions, respectively. Plots of $\log \tau$ vs $\frac{1}{T}$ should yield values of E_1 and E_2. If these plots yield straight lines then the implication is that $E_2 = 0$, and the non-radiative processes are not present. A consideration of figures III.4(a), 5(a) indicate that for alpha particle excitation of CsI(Tl) and CsBr(Tl), the plots of $\log \tau$ vs $\frac{1}{T}$ do in fact yield straight lines. We would then expect, however, the luminescence efficiency to be independent of temperature. This is clearly not the case. We must conclude

that the above model is unable to explain the observed experimental results on the luminescence of thallium activated alkali halide crystals. In any case the model makes no provision for the variation of γ with ionization density.

Two simple explanations for the observed variation of γ with ionization density could have been the following. Firstly, as suggested by Storey et al. (loc.cit.), protons and alpha particles may excite the same electron trap in the crystal. The probability for the decay of the trap is $\alpha^{at} \exp(\frac{E}{kT})$. Due to an increased effective temperature in the material surrounding the particle track, corresponding to the higher ionization density, the decay time for the alpha particles is faster. In this case, as the temperature of the crystal is lowered, the ratio $\gamma_{ip}/\gamma_{i\alpha}$ would increase. This is contrary to the observed behaviour shown in Figure III.8. A second explanation could have been that protons preferentially excite traps with greater activation energies. The decay time for protons would then be longer than for alpha particles. A consideration of the values of $E_{i\alpha}$ and E_{ip} found in CsI(Tl)(Chapter III Section 1(c)) indicates that this is not the case, E_{ip} is apparently smaller than $E_{i\alpha}$. For the alpha particle excitation of

CsBr(Tl), it again appears that the long components correspond to traps with smaller activation energies (Chapter III, Section 1(c)). Further evidence for this is afforded by a consideration of the ratios R_1, R_2 etc. given in Tables III.1(a),(b) and III.2(a),(b). In CsBr(Tl) it appears that a number of traps exist which decay radiatively when they receive enough thermal energy to reach an excited state, or to the trap immediately below in activation energies. At any one temperature, the trap responsible for the long component apparently feeds the trap responsible for the short component, the probability for such a transition increasing as the temperature of the crystal is lowered. Probably this is also the case in CsI(Tl). This observation that the larger values of τ correlate with smaller values of E, the activation energy, is a difficulty, but it does not negate a model in which electrons are released from traps. It does require, however, that the decay be governed by other factors apart from the activation energy of the electron traps, probably on the cross section for capture of electrons by the traps. Summarising, we can say that the observed variation of τ, the decay time of the luminescence, with ionization density, is not simply due to particles of increasing ionization

density exciting traps with greater activation energies, nor is it due to any local heating effect. In fact the decay time of the luminescence is not governed, apparently, solely by the activation energy of electron traps.

The variation in τ with ionization density could be explained, as suggested by Storey et al. (loc.cit.), if the variation in ionization density results in a different distribution in the initial population of radiating states. The observed variation in the value of τ would then reflect the different contributions of many radiating states with different decay times. The simplest and most extreme case would be two radiating states corresponding to the values of τ observed for alpha particles and electrons. This is the view taken by Ely and Jentschke (loc.cit.) and by Hrehuss (loc.cit.). The observed values of τ seem, however, to be very long to be associated with radiative states, particularly when the variation of τ with temperature is considered.

The only paper which has predicted quantitatively the behaviour of the luminescence is that by Meyer and Murray (loc.cit.) who have calculated, as we have seen in Chapter I, the response of CsI(Tl) as a function of thallium concentration and ionization density and obtain good agreement

with experiment. Their model interprets the response of inorganic crystals to charged particles in terms of the formation, transport, and capture of energy carriers. The basic assumption is that the energy carriers are excitons and that the density of the excitons is a smooth function of $\frac{dE}{dx}$, the ionization density. The model predicts that the response should show a maximum with ionization density in agreement with the results presented in Tables II.1-5. Again the model would predict that $^{\infty}\varepsilon_{\alpha}$ would increase very much more markedly with thallium concentration than $^{\infty}\varepsilon_{e}$. This again agrees with the experimental results shown in figure III.10. The only criticism of this paper is that the results are dependent on the diffusion length of the excitons in the crystal. This is obtained by fitting the theoretically predicted response curve to the experimentally obtained response curve. This is dependent on the spectral response of the photomultiplier used and hence if the emission spectra for proton excitation is different from that of alpha particle excitation the response obtained will not show the true relative efficiencies. However the success of the model indicates that this effect, if present, must be slight.
To explain the behaviour of $^{\infty}\varepsilon$ with temperature, it is necessary that the diffusion length be a function of

temperature and also, possibly, that the number of excitons created for a particular ionization density also shows a variation with temperature. Even so, it is difficult to understand why $^{\infty}\varepsilon_{\alpha}$ should become greater than $^{\infty}\varepsilon_{\rho}$, at low temperatures, as we find in both CsI(Tl) and CsBr(Tl). Mateosian et al. (loc.cit.) find a similar effect in pure NaI below -150°C. It should be noted that this model is in contradiction with the models of Johnson and Williams (loc. cit.) and Kallman and Brucher (loc.cit.) who predict that the luminescence efficiency should decrease for too large concentrations of thallium. This model predicts the response of the inorganic phosphors completely independently of the decay processes or the potential configurations of the luminescence centres.

The suggestion of Mateosian et al. (loc.cit.) that the behaviour of the luminescent efficiency $^{\infty}\varepsilon$ with temperature is the result of two different types of luminescence centre in the crystals, as discussed in Chapter I, can be discounted from the behaviour of the decay times of the luminescence with temperature. On this model we should expect to find a decay time component which becomes faster as the temperature is decreased.

IV.2. The Function of Thallium in the Luminescence Process.

As we have seen, the role of the thallium in the luminescence process is very far from clear. Our results for the behaviour of $^{\infty}\varepsilon_{\alpha}$ and $^{\infty}\varepsilon_{e}$ versus thallium concentration (Figure III.10) would indicate that, in agreement with Meyer and Murray (loc.cit.), and Ely and Jentschke (loc.cit.), the thallium is indeed the luminescence centre. Or it may be inducing the luminescence centres in the crystal as suggested by Knoepfel et al. (loc.cit.) and the results of Lwles and Joshi (loc.cit.). It is difficult to see how the thallium can be associated with the non-radiative processes in the crystal, as suggested by Bonanomi and Rossel (loc.cit.) since then the luminescent efficiency would be expected to decrease as the thallium concentration is increased. The situation becomes complicated however when one considers the behaviour of γ_{α} and γ_{e} with thallium concentration (figure III.9). Since γ_{e} remains constant with thallium concentration it would appear that the state from which the luminescence is emitted is unaffected by the thallium. A more suitable picture would seem to be, that for excitation of the crystal by particles of low ionization density, the luminescence emission is independent of the thallium. For particles of high ionization

density, at low thallium concentrations, the emission is again independent of the thallium, since $\tau_e = \tau_\alpha$ but that as the thallium concentration increases the emission becomes more dependent on the thallium. We then require two states from which luminescence can be emitted. The first state is populated preferentially by particles of low ionization density and is independent of thallium. The second is populated preferentially by particles of high ionization density and is induced by the thallium. The thallium state has the characteristic faster decay time. The difference in τ with ionization density and thallium concentration would then be due to the different probabilities of exciting these states.

IV.3. Conclusions.

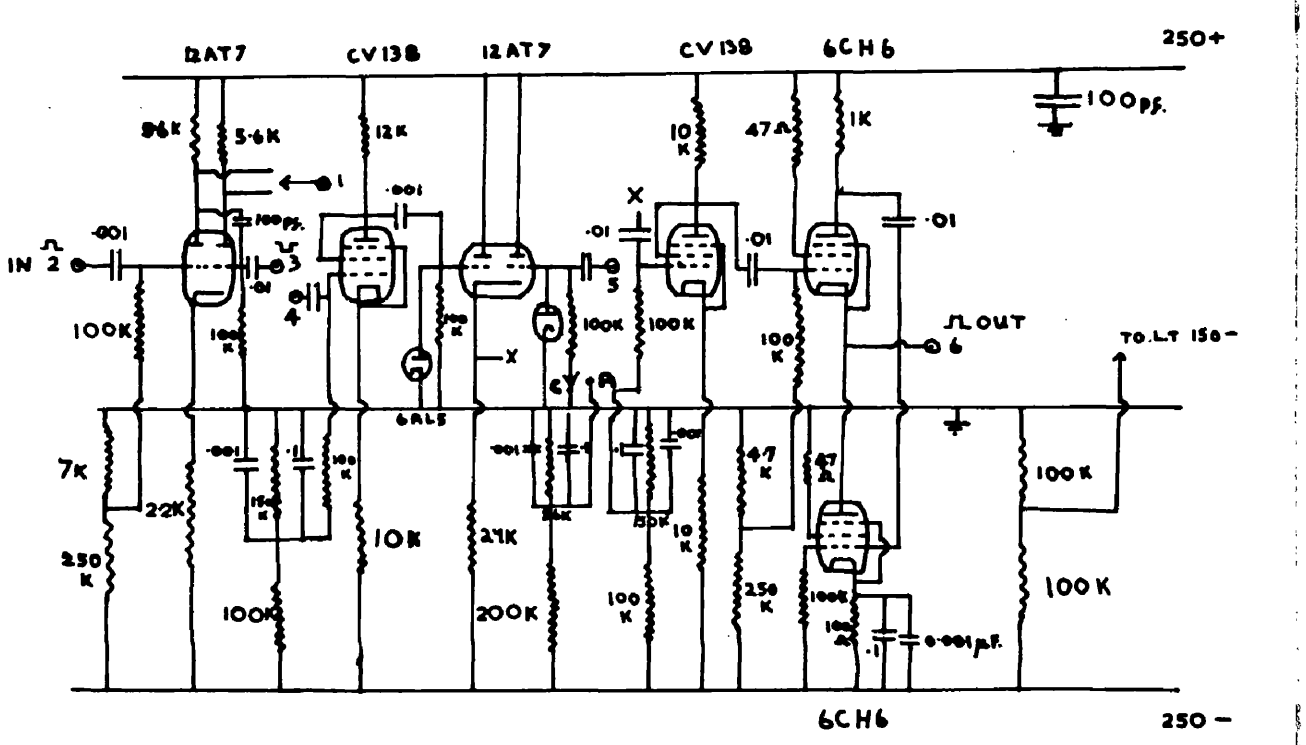
We must conclude that the measurements on the luminescence of the alkali halide crystals cannot be explained on the basis of any of the proposed models for the luminescence processes. All of these proposed models can account for some of the experimental observations but none can completely explain the dependence of the luminescence efficiency, and the decay times of the luminescence, on ionization density, temperature and thallium concentration.

The most likely approach would seem to be to take the view that energy carriers are formed by the exciting particles, the number of carriers being dependent on ionization density and to then consider the transport of these carriers to the luminescent centres. Meyer and Murray have produced in this way, as we have seen, the only quantitative prediction on the luminescence of the alkali halide phosphors. A detailed knowledge of the luminescence centres is then required. Probably this can best be obtained in the classical manner by exciting the crystals by light or ultra-violet radiation of a single known wavelength. In particle excitation of course a band of wavelengths is involved. It is probably fair to say that too much has been inferred about the luminescence processes from too few experimental observations.

Future Programme.

It is not intended to make any further measurements on the luminescence of inorganic alkali halide crystals, at present. The necessary information concerning the technique described in Part I has been obtained and it is intended to use this in nuclear physics measurements as discussed in Part I. Any future measurements would be made on the luminescence of pure alkali halides, again as a function of ionization density and temperature. In addition measurements would be made of the emission spectra of pure and activated crystals under both ultra-violet and particle excitation.

Appendix.



Circuit Diagram of coincidence-anti coincidence gate circuit used in the pulse shape discrimination technique.

References.

Ahn, Se. Hee., Hearst, J.R., Roberts, J.H. and Strait, E.N., 1960, Phys. Rev., 119, 1667.

Allan, D.L., 1957, Proc. Phys. Soc., A., 70, 195; 1958, Nuc. Phys., 6, 464; 1959, Ibid, 10, 348.

Allison, S.K., and Casson, H., 1953, Phys. Rev., 90, 880.

Armstrong, A.H., and Rosen, L., 1960, Nuc. Phys., 19, 40.

Austern, N, Butler, S.T., and McManus, H., 1953, Phys. Rev., 92, 350.

Austern, N., and Butler, S.T., 1958, Phys. Rev., 109, 1402.

Bame, S.J., Jr., Haddad, E., Perry, J.E., Jr., and Smith, R.K., 1957, Rev. Sci. Instr., 28, 997.

Becker, R.L., 1960, Phys. Rev., 119, 1076.

Biggerstaff, J.A., and Becker, R.L., 1959, Bull. Am. Phys. Soc., 4, 326.

Bittman, M., Furst, M., and Kallman, H., 1952, Phys. Rev., 87, 83.

Blatt, J.M. and Weisskopf, V.F., 1952, 'Theoretical Nuclear Physics', (New York; Wiley).

Blau, M., and Dreyfuss, B., 1945, Rev. Sci., Instr., 16, 245.

Bonanomi, J., and Rossel, J., 1952, Helv. Phys. Acta., 25, 725.

Brooks, F.D., 1959, Nuclear Instr. and Methods, 4, 151.

Brown, G., and Muirhead, H., 1957, Phil. Mag., 2, 473.

Butler, S.T., Austern, N., and Pearson, C., 1958, Phys. Rev., 112, 1227.

Chauvy, D., and Rossel, J., 1959, Helv. Phys. Acta., 32, 481.

Cohen, B.L., 1953, Phys. Rev., 92, 1245.

Coleman, R.F., Hawker, B.E., O'Conner, L.P., and Perkin, J.L., 1959, Proc. Phys. Soc., 73, 215.

Colli, L., Facchini, U., 1956, Nuovo Cim., 4, 671.

Colli, L., Facchini, U., Micheletti, S., 1957, Nuovo Cim., 5, 502.

Curran, S.C., 1953, 'Luminescence and the Scintillation Counter', (London, Butterworth's Scientific Publications).

Curran, S.C., and Baker, W., Rad. Lab. Rep. Berkeley 7-6-16, Nov. 17, 1944.

Dexter, D.L., 1953, J. Chem. Phys. 21, 836.

Dosch, H.G., Finckh, E., Lindenberger, K.H., Mailer, K. and Meyer-Berkhout, 1960, Proc. Karlsruhe Conference.

Eby, F.S. and Jentschke, W.K., 1954, Phys. Rev., 96, 911.

Elton, L.R.B., and Gomes, L.C., 1957, Phys. Rev., 105, 1027.

Enz, H. and Rossel, J., 1959, Helv. Phys. Acta., 31, 25.

Eubank, H.P., Peck, R.A., and Hassler, F.L., 1958/59, Nuc. Phys., 9, 273.

Eubank, H.P., Peck, R.A., Jr., and Zatzick, M.R., 1959, Nuc. Phys., 10, 418.

Ewles, J., and Joshi, R.V., 1960, Proc. Royal Soc., A., 254, 358.

Fleury, J., Perrin, P., Boge, M., and Laugier, J., 1960, J. De Physique et Le Radium, 21, 480

Franzen, W., Peelo, R.W., and Scherr, R., 1950, Phys. Rev., 79, 742.

Fulmer, C.B., and Cohen, B.L., 1958, Phys. Rev., 112, 1672.

Garlick, G.F.J., 1952., Prog. in Nuclear Physics, 2, 51.

Glendenning, N.K., 1959, Phys. Rev., 114, 1297.

Gugelot, P.C., 1954, Phys. Rev., 93, 425.

Hahn, B., and Rossel, J., 1953a, Helv. Phys. Acta., 26, 271; 1953b, Ibid, 26, 803.

Haling, R.K., Peck, R.A., and Eubank, H.P., 1957, Phys. Rev., 106, 911.

Hayakawa, S., Kawai, M. and Kickuchi, K., 1955, Prog. Theor. Phys., 13, 415.

Heppel, A. Von, 1936, Z. Phys., 101, 680.

Hofstadter, R., 1948, Phys. Rev., 74, 100, 628.

Hutchinson, G.W., and Scarrott, G.G., 1951, Phil. Mag., 42, 792.

Hrehuss, G., 1960, Nuclear Inst. and Methods, 8, 344.

Jack, W., and Ward, A., 1960, Proc. Phys. Soc., 75, 833.

Johnson, P.D., and Williams, F.E., 1950, J. Chem. Phys., 18, 1477.

Jones, D.A., and Ward, A., 1960, Proc. Phys. Soc., 75, 931.

Kallman, H., and Brucker, G.J., 1957, Phys. Rev., 108, 1122.

Kallman, H., and Furst, M., 1950, Phys. Rev., 79, 857.

Kienle, P., and Segel, R.E., 1959, Phys. Rev., 113, 909.

Knoepfel, H., Loepfe, E., and Stoll, P., 1957, Helv. Phys. Acta., 30, 521.

Knox, R.S., and Dexter, D.L., 1956, Phys. Rev., 104, 1245.

Kumabe, I., Takekoshi, E., Ogata, H., Tsunoda, Y., and Oki, S., 1957, Phys. Rev., 106, 155.

March, P.V., and Morton, W.T., 1958a, Phil. Mag. 3, 143;
1958b, Ibid, 3, 577;
1958c, Ibid, 3, 1256.

Mateosian, E. Der., McKeown, M., Muehlhause, C.O., 1956, Phys. Rev., 101, 967.

Meyer, A., and Murray, R.D., 1960, Nucleonics, 18, 86.

McManus, H., and Sharpe, W.T., 1952, Phys. Rev., 87, 188.

Nicholson, K.P., and Snelling, G.F., 1955, British J. Appl. Phys., 6, 104.

Owen, R.B., 1959, Nucleonics, 17, 92.

Paul, E.B., and Clarke, R.L., 1953, Canad. J. Phys., 31, 267.

Peck, R.A., Jr., 1957, Phys. Rev., 106, 965.

Randall, J.T., and Wilkins, M.H.F., 1945, Proc. Royal Soc., 184, 366.

Seitz, F., 1938, J. Chem. Phys., 6, 150.

Seitz, F., 1939, Trans. Faraday Soc., 35, 74.

Scharmann, A., 1959, Zeitschrift fur Physik, 157, 301.

Schenk, J. and Neiler, J.H., 1954, Nucleonics, 12, 28.

Seiver, Van W.J., 1956, Nucleonics, 14, 50.

Stockbarger, D.C., 1949, J. Opt. Soc. Amer., 39, 731.

Storey, R.S., Jack, W., and Ward, A., 1958, Proc. Phys. Soc. 72, 1.

Storey, R.S., Jack, W., and Ward, A., 1960, Proc. Phys. Soc., 75, 526.

Swank, R.K., 1954, Annual Review Nuclear Science, 4, 111.

Thomson, D.H., 1956, Proc. Phys. Soc., A., 69, 447.

Weisskopf, V.F., 1937, Phys. Rev., 52, 295.

Whetstone, A., Allison, B., Muirhead, D.G., and Halpern, J., 1958, Rev. Sci. Instr. 29, 415.

Wilkinson, D.H., 1952, Rev. Sci. Instr., 23, 414.

Williams, F.E., 1951, J. Chem. Phys., 19, 457; 1953, Ibid, 57, 780.

Wright, G.T., 1953, Phys. Rev., 91, 1282.